



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Manabu Tsumura et al.

Serial No.: 10/512,135

Art Unit: 1712

Filed: May 3, 2005

Examiner: Marc S. Zimmer

Title: HARDENABLE COMPOSITION, HARDENING PRODUCT, PROCESS
FOR PRODUCING THE SAME AND LIGHT EMITTING DIODE
SEALED WITH THE HARDENING PRODUCT

DECLARATION UNDER RULE 1.132

Honorable Commissioner of Patents and Trademarks,
Alexandria, Virginia 22313-1450

Sir:

I, Masahito Ide, a citizen of Japan and having postal mailing address of 5-5-35-210, Torikai-nishi, Settsu-shi, Osaka 566-0072 JAPAN, declare and say that:

March, 2001, I was graduated from Osaka University and received a Master Degree in Graduate School of Engineering Science;

Since April, 2001, I have been employed by Kaneka Corporation, and engaged in the works of research and development for elastomer materials at High Performance Materials Research Laboratories in Corporate Research and Development Division;

I have read the Official Action mailed and the references cited therein and I am familiar with the subject matter thereof;

I respectfully submit herewith my exact report thereon;

Experiments

The following experiments were conducted.

Experiment 1

(Synthesis Example 1')

A 2-L autoclave was charged with 602 g of toluene and 626 g of 1,3,5,7-tetramethylcyclotetrasiloxane, and the atmosphere was substituted by nitrogen gas. The mixture was heated at 105°C of the inside temperature and stirred. Thereto was added dropwise a mixture of 90.0 g of triallyl isocyanurate, 0.057 g of platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) and 90.0 g of toluene in 40 minutes. After 4 hours from the end of dropwise addition, not less than 95% of reaction rate of allyl group was observed by ^1H -NMR, and the reaction was finalized by cooling. The unreacted portion of 1,3,5,7-tetramethylcyclotetrasiloxane and the toluene were distilled off under reduced pressure. ^1H -NMR revealed that the product was the reaction product resulting from part of the SiH groups of 1,3,5,7-tetramethylcyclotetrasiloxane with allyl group of triallyl isocyanurate (hereinafter referred to as partial reaction product B1', SiH value: 9.21 mmol/g, allyl value: 0.10 mmol/g).

(Example X)

Triallyl isocyanurate (17.29 g), 27.60 g of diallyl monoglycidyl isocyanurate (61% by weight in component (A)) and 290 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) were mixed up to prepare a mixture A. Separately, a mixture B was prepared in advance by mixing up 51.64 g of

the partial reaction product (B1') prepared in Synthesis Example 1' and 290 mg of 1-ethynyl-1-cyclohexanol. A one-pot mixture was prepared by mixing up the above mixtures A and B with stirring and degassing. The rate of SiH/allyl in the one-pot mixture was 1.14. The above one-pot mixture was poured into a cell produced by inserting a 3-mm-thick silicone rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour, 150°C for 1 hour, and 180°C for 1 hour, in that order, to obtain a transparent and hard molding.

(Comparative Example X)

Triallyl isocyanurate (38.75 g) and 290 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) were mixed up to prepare a mixture A. Separately, a mixture B was prepared in advance by mixing up 57.77 g of the partial reaction product (B1') prepared in Synthesis Example 1' and 290 mg of 1-ethynyl-1-cyclohexanol. A one-pot mixture was prepared by mixing up the above mixtures A and B with stirring and degassing. The rate of SiH/allyl in the one-pot mixture was 1.14. The above one-pot mixture was poured into a cell produced by inserting a 3-mm-thick silicone rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour, 150°C for 1 hour, and 180°C for 1 hour, in that order, to obtain a transparent and hard molding.

(Measurement Example I)

For the samples obtained in Example X and Comparative Example X, flexural strength and flexural

elongation were evaluated. The test samples were prepared as follows. The plate-shaped cured products of 3 mm in thickness were cut into rectangles of 700×50×3 mm. The measurement was carried out using an autograph placed under the condition of 50% RH at 23°C, and the distance between the fulcrums was set at 50 mm. The flexural test was carried out at a speed of 1.5 mm/min. The obtained results are shown in Table I.

Table I

	Strength (N)	Elongation (%)
Example X	66	4.7
Comparative Example X	65	3.7

From Table I, it is found that the cured product of the curable composition of Example X, which contains a compound (II) in an amount of 20% by weight or more in the component (A), has higher strength and elongation as well as higher toughness than the cured product of the curable composition of Comparative Example X, which does not contain a compound (II) in an amount of 20% by weight or more in the component (A).

Experiment 2

(Synthesis Example 2')

A 2-L autoclave was charged with 362 g of toluene and 362 g of 1,3,5,7-tetramethylcyclotetrasiloxane, and the atmosphere was substituted by nitrogen gas. The mixture was heated at 105°C of the inside temperature and stirred. Thereto was added dropwise a mixture of 100.0 g of diallyl monoglycidyl isocyanurate, 0.049 g of platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) and 100.0 g of

toluene in 40 minutes. After 4 hours from the end of dropwise addition, not less than 95% of reaction rate of allyl group was observed by $^1\text{H-NMR}$, and the reaction was finalized by cooling. The unreacted portion of 1,3,5,7-tetramethylcyclotetrasiloxane and the toluene were distilled off under reduced pressure. $^1\text{H-NMR}$ revealed that the product was the reaction product resulting from part of the SiH groups of 1,3,5,7-tetramethylcyclotetrasiloxane with allyl group of diallyl monoglycidyl isocyanurate (hereinafter referred to as partial reaction product B2', SiH value: 7.74 mmol/g, allyl value: 0.10 mmol/g).

(Example Y)

Triallyl isocyanurate (35.63 g) and 290 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) were mixed up to prepare a mixture A. Separately, a mixture B was prepared in advance by mixing up 60.89 g of the partial reaction product (B2') prepared in Synthesis Example 2' and 290 mg of 1-ethynyl-1-cyclohexanol. A one-pot mixture was prepared by mixing up the above mixtures A and B with stirring and degassing. The rate of SiH/allyl in the one-pot mixture was 1.1. The above one-pot mixture was poured into a cell produced by inserting a 3-mm-thick silicone rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour, 150°C for 1 hour, and 180°C for 1 hour, in that order, to obtain a transparent and hard molding.

(Comparative Example Y)

Triallyl isocyanurate (39.59 g) and 290 mg of a

platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) were mixed up to prepare a mixture A. Separately, a mixture B was prepared in advance by mixing up 56.93 g of the partial reaction product (B1') prepared in Synthesis Example 1' and 290 mg of 1-ethynyl-1-cyclohexanol. A one-pot mixture was prepared by mixing up the above mixtures A and B with stirring and degassing. The rate of SiH/allyl in the one-pot mixture was 1.1. The above one-pot mixture was poured into a cell produced by inserting a 3-mm-thick silicone rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour, 150°C for 1 hour, and 180°C for 1 hour, in that order, to obtain a transparent and hard molding.

(Measurement Example II)

For the samples obtained in Example Y and Comparative Example Y, flexural strength and flexural elongation were evaluated. The test samples were prepared as follows. The plate-shaped cured products of 3 mm in thickness were cut into rectangles of 700×50×3 mm. The measurement was carried out using an autograph placed under the condition of 50% RH at 23°C, and the distance between the fulcrums was set at 50 mm. The flexural test was carried out at a speed of 1.5 mm/min. The obtained results are shown in Table II.

Table II

	Strength (N)	Elongation (%)
Example Y	73	4.4
Comparative Example Y	65	3.7

From Table II, it is found that the cured product of the

curable composition of Example Y, in which the component (B) contains a reaction product from 1,3,5,7-tetramethylcyclotetrasiloxane and diallyl monoglycidyl isocyanurate, and/or a reaction product from 1,3,5,7-tetramethylcyclotetrasiloxane and monoallyl diglycidyl isocyanurate, has higher strength and elongation as well as higher toughness than the cured product of the curable composition of Comparative Example Y, which does not contain the above reaction product in the component (B).

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this day of May, 2007

May 18, 2007

Masahito Ide

Masahito Ide

VERIFICATION OF TRANSLATION



Fumio Akiyama of MT-2 BLDG., 5-36, Miyahara 3-chome, Yodogawa-ku, Osaka-shi, Osaka 532-0003 JAPAN, hereby declare that I am conversant with the Japanese and English languages and that I am the translator of the documents attached and certify that to the best of my knowledge and belief the following is a true and correct English translation of the Japanese Patent Application No. 2002-133412 in the name of KANEKA CORPORATION.

Dated this 16th day of May, 2007

Fumio Akiyama

Japanese Patent Application No.2002-133412 Filed on May 9, 2002

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[Document Name] Patent Application
[Filing Number] OSK-4764
[To] Commissioner, Patent Office
[Inventor]
[Address or Residence] A101, Kozenryo, 2-23, Torikainishi
5-chome, Settsu-shi, Osaka
[Name] TUMURA Manabu
[Inventor]
[Address or Residence] D202, Kozenryo, 2-23, Torikainishi
5-chome, Settsu-shi, Osaka
[Name] IDE, Masahito
[Inventor]
[Address or Residence] 5-35-505, Torikainishi 5-chome,
Settsu-shi, Osaka
[Name] OUCHI, Katsuya
[Applicant]
[Identification Number] 000000941
[Name] KANEKA CORPORATION
[Representative] TAKEDA Masatoshi
[Indication of Fee]
[Number of Deposit Ledger] 005027
[The Amount of Payment] 21,000 yen
[List of Attached Documents]
[Document Name] Description 1
[Document Name] Abstract 1
[Necessity of Proof] Needed

[Document Name] Specification

[Title of the Invention] HARDENABLE COMPOSITION, HARDENING PRODUCT, PROCESS FOR PRODUCING THE SAME AND LIGHT EMITTING DIODE SEALED WITH THE HARDENING PRODUCT

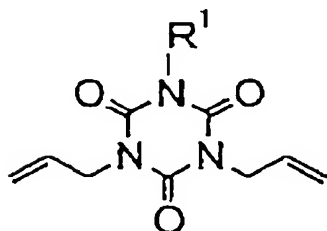
5 [Scope of Claims for Patent]

[Claim 1] A curable composition which contains

(A) a compound represented by the general formula (I):

[Chemical 1]

10



(I)

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in the formula, R¹ represents an organic group which does not contain a functional group subjectable to hydrosilylation reaction,

(B) a silicon compound having at least two SiH groups in each molecule, and

20

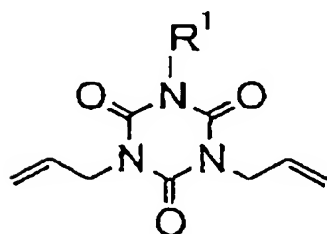
(C) a hydrosilylation catalyst.

[Claim 2] A curable composition which contains

(A) triallyl isocyanurate, a compound represented by the general formula (I):

[Chemical 2]

25



(I)

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in the formula, R¹ represents an organic group which does not contain a functional group subjectable to hydrosilylation reaction,

(B) a silicon compound having at least two SiH groups in each molecule, and

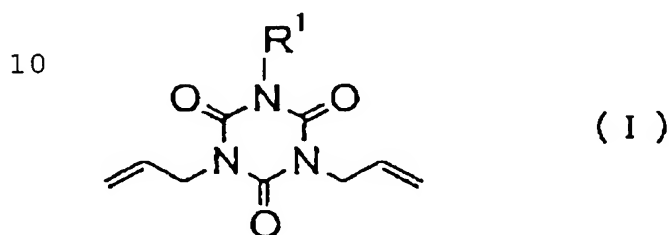
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(C) a hydrosilylation catalyst.

[Claim 3] The curable composition according to Claim 1 or 2,
wherein R^1 represents a hydrogen atom, or a univalent
organic group containing 1 to 50 carbon atoms.

5 [Claim 4] The curable composition according to Claim 2,
wherein the compound represented by the general formula
(I):

[Chemical 3]



15 in the formula, R^1 represents an organic group which does
not contain a functional group subjectable to hydrosilylation
reaction, accounts for 20% by weight or more in the component
(A).

[Claim 5] The curable composition according to Claim 2,
20 wherein the component (A) is triallyl isocyanurate and
diallyl monoglycidyl isocyanurate.

[Claim 6] The curable composition according to Claim 2,
wherein the component (A) is triallyl isocyanurate and
diallyl monoglycidyl isocyanurate and the component (B) is a
25 reaction product from 1,3,5,7-tetramethylcyclotetrasiloxane
and triallyl isocyanurate.

[Claim 7] A curing product

which is obtainable by curing the curable composition
according to any one of Claims 1 to 6.

30 [Claim 8] A process for producing a curing product

which comprises curing the curable composition according
to any one of Claims 1 to 6.

[Claim 9] A light-emitting diode

35 7. which is sealed with the curing product according to Claim

[Detailed Description of the Invention]

[0001]

[Technical Field of the Invention]

The present invention relates to a curable composition
5 for optical materials, more specifically a curable composition
for optical materials having high toughness and good light
resistance, a process for producing the same, a curing product
thereof, and a light-emitting diode sealed with the curing
product.

10 [0002]

[Prior Art]

A curable composition is known which comprises an
aliphatic organic compound having, within the molecule, at
least two carbon-carbon double bonds reactive with a SiH group,
15 a compound containing at least two SiH groups within the
molecule, and a hydrosilylation catalyst (WO 01-81475). A
curing product obtainable by curing this curable composition
can be preferably used as an optical material. However, in
applications for films as optical materials, e.g. molded
20 products such as lens, or sealing materials, high mechanical
properties are demanded in addition to the optical
characteristics in view of long-term reliability of materials.
Herein, when the crosslinking density is high, the material
becomes fragile, thus a problem of decrease in tensile
25 characteristics arose.

[0003]

[Subject which the Invention is to Solve]

Accordingly, the object of the present invention is to
provide a curable composition having high toughness and good
30 light resistance, a process for producing the same, a curing
product thereof, and a light-emitting diode sealed with the
curing product.

[0004]

[Means for Solving the Problems]

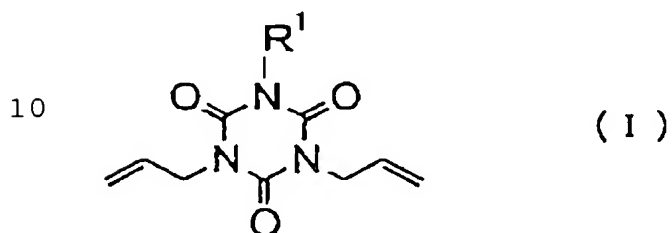
35 The present inventors made intensive investigations to

accomplish the above object and, as a result, found that it is possible to obtain a curable composition having high toughness and good light resistance by using a curable composition which contains

5 (A) a compound represented by the general formula (I):

[0005]

[Chemical 4]



15 in the formula, R¹ represents an organic group which does not contain a functional group subjectable to hydrosilylation reaction,

(B) a silicon compound having at least two SiH groups in each molecule, and

(C) a hydrosilylation catalyst.

20 Based on this finding, they have now completed the present invention.

[0005]

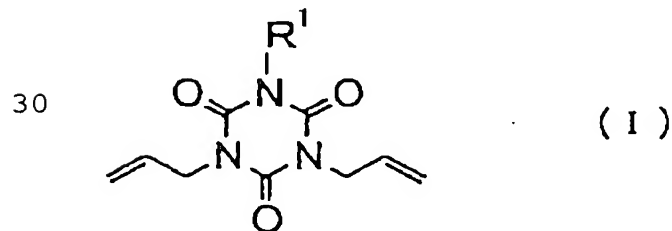
Thus, the present invention is:

A curable composition which contains

25 (A) a compound represented by the general formula (I):

[0006]

[Chemical 5]



35 in the formula, R¹ represents an organic group which does not contain a functional group subjectable to hydrosilylation

reaction,

(B) a silicon compound having at least two SiH groups in each molecule, and

(C) a hydrosilylation catalyst (Claim 1),

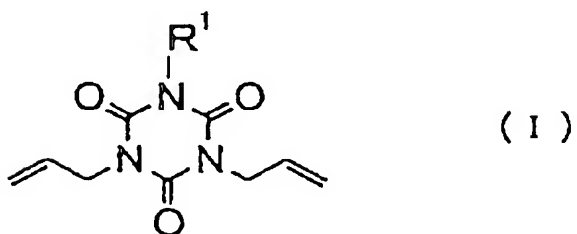
5 A curable composition which contains

(A) triallyl isocyanurate, a compound represented by the general formula (I):

[0007]

[Chemical 6]

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15

in the formula, R¹ represents an organic group which does not contain a functional group subjectable to hydrosilylation reaction,

20 (B) a silicon compound having at least two SiH groups in each molecule, and

(C) a hydrosilylation catalyst (Claim 2),

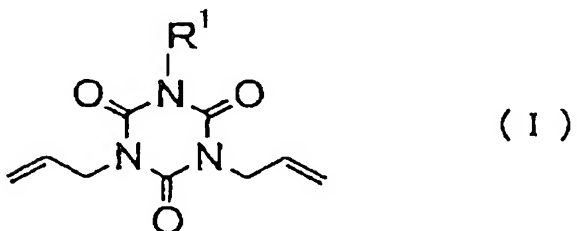
The curable composition according to Claim 1 or 2, wherein R¹ represents a hydrogen atom, or a univalent organic group containing 1 to 50 carbon atoms (Claim 3),

25 The curable composition according to Claim 2, wherein the compound represented by the general formula (I):

[0008]

[Chemical 7]

30



35

in the formula, R^1 represents an organic group which does not contain a functional group subjectable to hydrosilylation reaction, accounts for 20% by weight or more in the component (A) (Claim 4),

5 The curable composition according to Claim 2,
 wherein the component (A) is triallyl isocyanurate and diallyl monoglycidyl isocyanurate (Claim 5),

 The curable composition according to Claim 2,
 wherein the component (A) is triallyl isocyanurate and
10 diallyl monoglycidyl isocyanurate and the component (B) is a reaction product from 1,3,5,7-tetramethylcyclotetrasiloxane and triallyl isocyanurate (Claim 6),

 A curing product
 which is obtainable by curing the curable composition
15 according to any one of Claims 1 to 6 (Claim 7),

 A process for producing a curing product
 which comprises curing the curable composition according to any one of Claims 1 to 6 (Claim 8),

 A light-emitting diode
20 which is sealed with the curing product according to Claim 7 (Claim 9),

[0009]

[Embodiment of carrying out the Invention]

25 In the following, the present invention is described in detail.

[0010]

 First, the component (A) of the present invention is described.

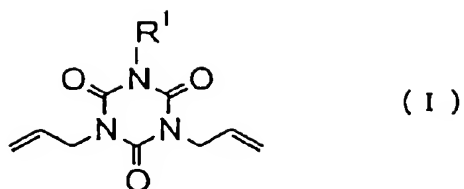
[0011]

30 The compound component (A) represented by the following general formula (I), which is characteristic for the present invention, is

[0012].

[Chemical 8]

35



5

in the formula, R¹ represents an organic group which does not contain a functional group subjectable to hydrosilylation reaction.

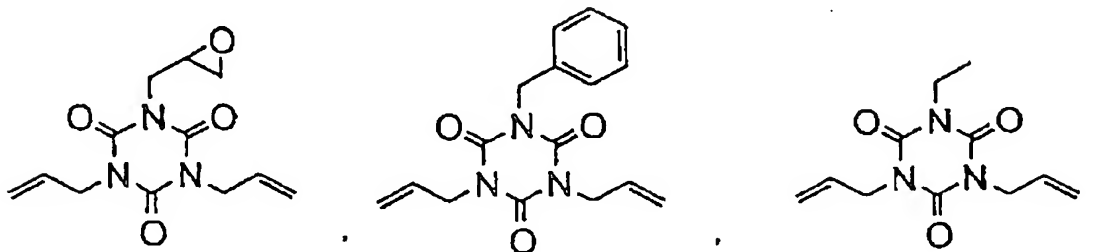
[0013]

10 As preferable example of these, there may be mentioned the following compounds:

[0014]

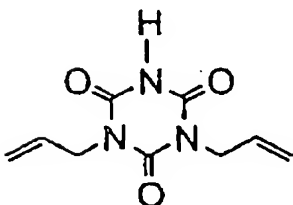
[Chemical 9]

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Among these, from the viewpoint that the mechanical and adhesive properties can be both obtained, diallyl monoglycidyl isocyanurate is particularly preferred.

The component (A) to be used may contain one single species or a mixture of two or more species. The component (A) which may be mixed together to be used is described.

35

[0015]

The above organic compound is preferably a compound other than a polysiloxane-organic block copolymer, a polysiloxane-organic graft copolymer or a like siloxane unit (Si-O-Si)-containing one, and a compound comprising C, H, N, O, S and/or halogen, as exclusive constituent elements. Siloxane unit-containing compounds have gas permeability and cissing problems.

[0016]

The sites of occurrence of the carbon-carbon double bonds reactive with a SiH group are not particularly restricted but may be anywhere within the molecule.

[0017]

The component (A) compound may be classified as an organic polymer type compound or as an organic monomer type compound.

[0018]

As the organic polymer type compounds, there may be mentioned, for example, polyether type, polyester type, polyarylate type, polycarbonate type, saturated hydrocarbon type, unsaturated hydrocarbon type, polyacrylic ester type, polyamide type, phenol-formaldehyde type (phenol resin type), and polyimide type compounds, etc.

[0019]

As the organic monomer type, there may be mentioned, for example, phenol type, bisphenol type, benzene, naphthalene or like aromatic hydrocarbon type compounds; aliphatic (e.g. straight chain, alicyclic) hydrocarbon type compounds; heterocyclic type compounds; mixtures of these, etc.

[0020]

The carbon-carbon double bonds reactive with a SiH group in the component (A) are not particularly restricted but, from the reactivity viewpoint, those groups represented by the following general formula (II):

[0021]

[Chemical 10]

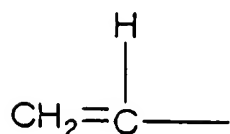


5 wherein R^2 represents a hydrogen atom or a methyl group, are preferred. From the material availability viewpoint, the group represented by

[0022]

[Chemical 11]

10



is particularly preferred.

15

[0023]

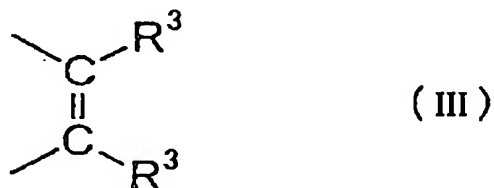
Moreover, preferred as the carbon-carbon double bonds reactive with a SiH group in the component (A) are alicyclic groups having a partial structure represented by the following general formula (III):

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[0024]

[Chemical 12]

25



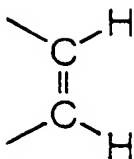
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wherein each R^3 represents a hydrogen atom or a methyl group, in the ring structure, since high heat resistance can be given to curing products. Among them, from the material availability viewpoint, alicyclic groups having a partial structure represented by the following formula

[0025]

[Chemical 13]

35

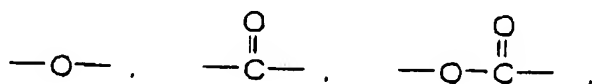


5 in the ring structure are preferred.

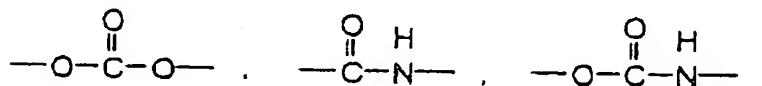
The carbon-carbon double bonds reactive with a SiH group each may be directly bonded to the skeletal portion of the component (A) or covalently bonded thereto via a bivalent or polyvalent substituent. The above bivalent or polyvalent
 10 substituent is not particularly restricted but preferably a substituent containing 0 to 10 carbon atoms, and more preferably one containing C, H, N, O, S and halogen, as exclusive constituent elements. Examples of the above-mentioned substituent are:

15 [0026]

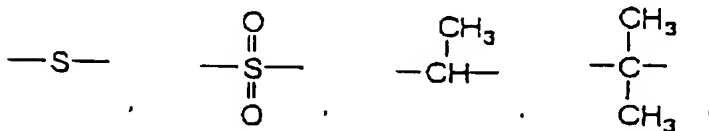
[Chemical 14]



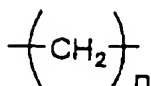
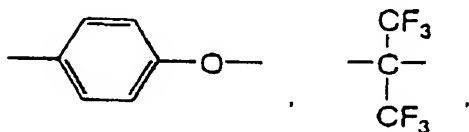
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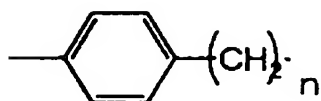


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(n representing a number of 1 to 10),

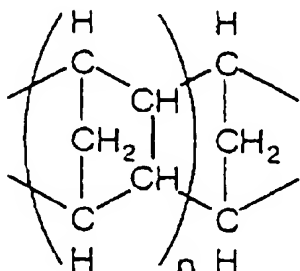


5 (n representing a number of 0 to 4), and

[0027]

[Chemical 15]

10



15 (n representing a number of 0 to 4), etc. Two or more of the bivalent or polyvalent substituents recited above may be covalently bonded together to form one bivalent or polyvalent substituent.

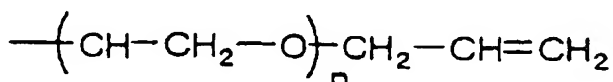
[0028]

20 As examples of the group covalently bonded to the skeletal portion, there may be mentioned vinyl, allyl, methallyl, acryl, methacryl, 2-hydroxy-3-(allyloxy)propyl, 2-allylphenyl, 3-allylphenyl, 4-allylphenyl, 2-(allyloxy)phenyl, 3-(allyloxy)phenyl, 4-(allyloxy)phenyl, 2-(allyloxy)ethyl, 25 2,2-bis(allyloxymethyl)butyl, 3-allyloxy-2,2-bis(allyloxymethyl)propyl,

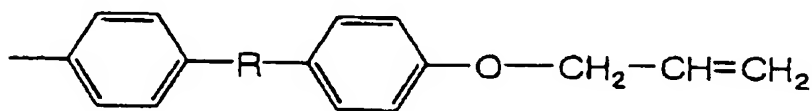
[0029]

[Chemical 16]

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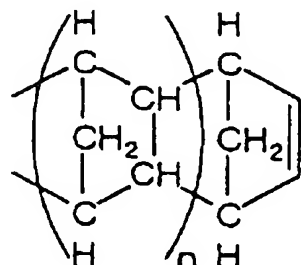
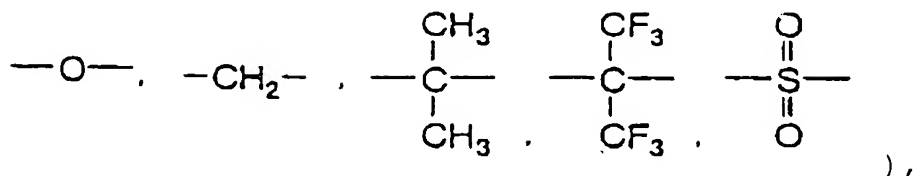


(n representing a number satisfying the relation $5 \geq n \geq 2$),



35

(R representing a bivalent group selected from among



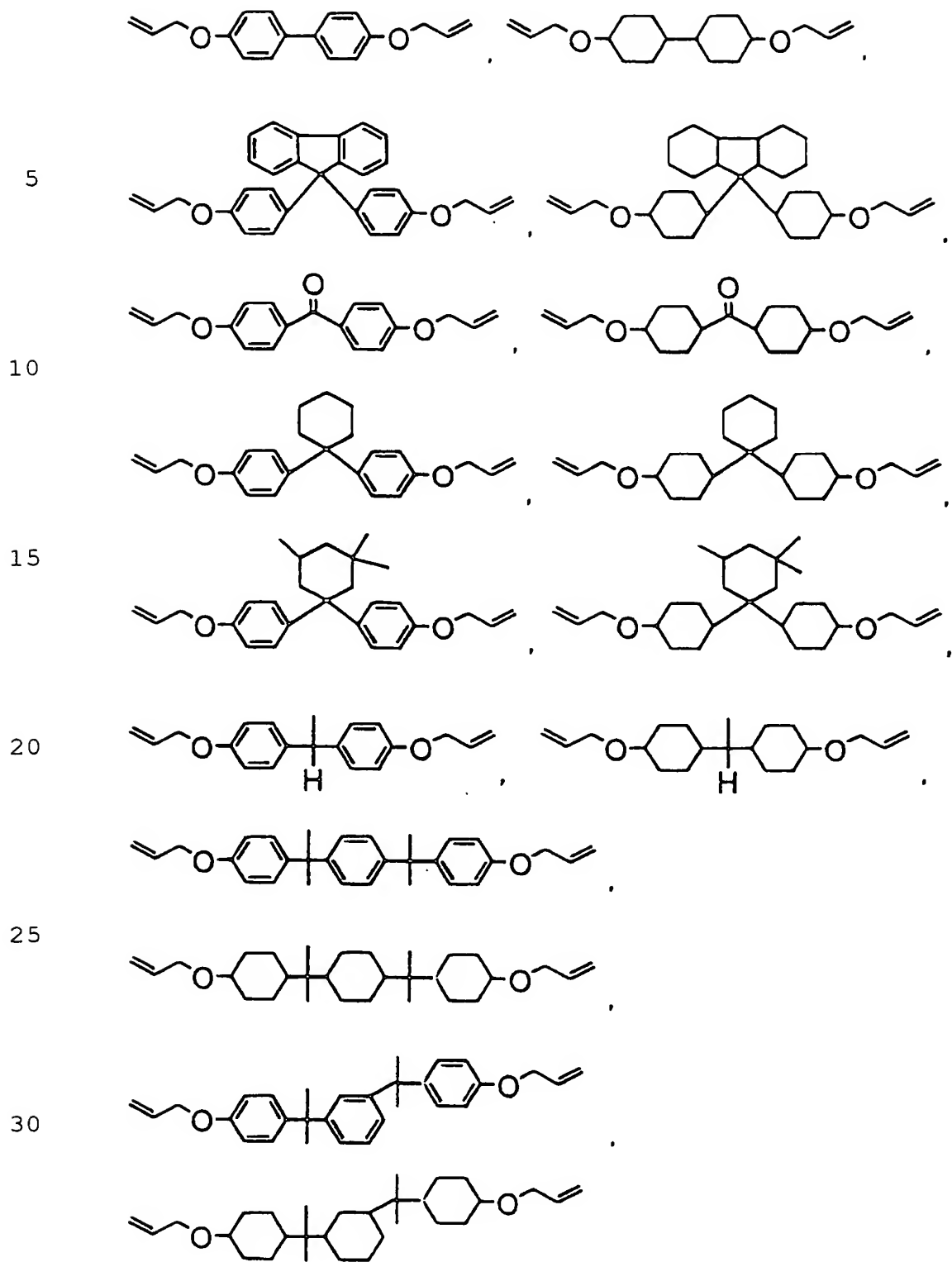
(n representing a number of 0 to 4), and the like.

[0030]

As specific examples of the component (A) which may be mixed together to be used, there may be mentioned diallyl phthalate, triallyl trimellitate, diethylene glycol bis(allyl carbonate), trimethylolpropane diallyl ether, pentaerythritol triallyl ether, 1,1,2,2-tetraallyloxyethane, diallylidenepentaerythritol, triallyl cyanurate, triallyl isocyanurate, 2,2-bis(4-hydroxy cyclohexyl)propane diallyl ether, 1,2,4-trivinylcyclohexane, divinylbenzenes (purity 50 to 100%, preferably 80 to 100%), divinylbiphenyl, 1,3-diisopropenylbenzene, 1,4-diisopropenylbenzene, and oligomers thereof, 1,2-polybutadiene (1,2 content 10 to 100%, preferably 50 to 100%), novolak phenol allyl ether, allylated poly(phenylene oxide),

[0031]

[Chemical 17]



35 and, further, products derived from known epoxy resins by allyl

group substitution for part or all of the glycidyl groups of the resins.

[0033]

Also usable as the component (A) which may be mixed
5 together to be used are low-molecular-weight compounds which can hardly be expressed dividedly in terms of a skeletal portion and an alkenyl group as mentioned above. As specific examples of the above low-molecular-weight compounds, there may be mentioned aliphatic linear polyene compounds such as butadiene,
10 isoprene, octadiene and decadiene, alicyclic polyene compounds such as cyclopentadiene, cyclohexadiene, cyclooctadiene, dicyclopentadiene, tricyclopentadiene and norbornadiene, substituted alicyclic olefin compounds such as vinylcyclopentene and vinylcyclohexene, etc.

15 [0034]

From the viewpoint of the possibility of achieving further improvements in heat resistance, it is preferred that the component (A) contain the carbon-carbon double bonds reactive with a SiH group in an amount of not less than 0.001
20 mole, more preferably not less than 0.005 mole, still more preferably not less than 0.008 mole, per gram of the component (A).

[0035]

As for the number of carbon-carbon double bonds reactive
25 with a SiH group in the component (A), it is only required that the number be at least 2 per molecule on average. For achieving further improvements in mechanical strength, the number is preferably more than 2, more preferably not less than 3. If the number of carbon-carbon double bonds reactive with a SiH
30 group in the component (A) is 1 or less per molecule, the component (A), upon reacting with the component (B), will only give a graft structure but will fail to give a crosslinked structure.

[0036]

35 From the good reactivity viewpoint, the component (A)

preferably contains at least one vinyl group, more preferably two or more vinyl groups, in each molecule. From the viewpoint of tendency toward better storage stability, it preferably contains not more than 6 vinyl groups, more preferably not more than 4 vinyl groups, in each molecule.

[0037]

From the viewpoint of high mechanical heat resistance and from the viewpoint of less stringiness, good moldability, and handleability, the component (A) preferably has a molecular weight lower than 900, more preferably lower than 700, still more preferably lower than 500.

[0038]

For attaining uniform admixture with other components and good workability, the component (A) preferably has a viscosity at 23°C of lower than 1000 P, more preferably lower than 300 P, still more preferably lower than 30 P. The viscosity can be determined using an E type viscometer.

[0039]

From the viewpoint of inhibition of coloration (in particular yellowing), the component (A) is preferably low in content of a compound having a phenolic hydroxyl group and/or a group derived from a phenolic hydroxyl group and, more preferably, it is free of any compound having a phenolic hydroxyl group and/or a group derived from a phenolic hydroxyl group. The term "phenolic hydroxyl group" so referred to herein means a hydroxyl group directly bound to an aromatic hydrocarbon moiety, for example a benzene, naphthalene or anthracene ring, and the "group derived from a phenolic hydroxyl group" means a group resulting from substitution of an alkyl (e.g. methyl, ethyl, etc.), alkenyl (e.g. vinyl, allyl, etc.), acyl (e.g. acetoxyl, etc.) group for the hydrogen atom of the above phenolic hydroxyl group.

[0040]

From the viewpoint of preferable optical characteristics such as low complex reflective index and low optical elastic

coefficient as well as preferable weather resistance, the component (A) preferably contains the aromatic ring component weight ratio of not more than 50% by weight, more preferably 40% by weight, and still more preferably 30% by weight. Most
 5 preferably, the component (A) does not contain any aromatic hydrocarbon ring.

[0041]

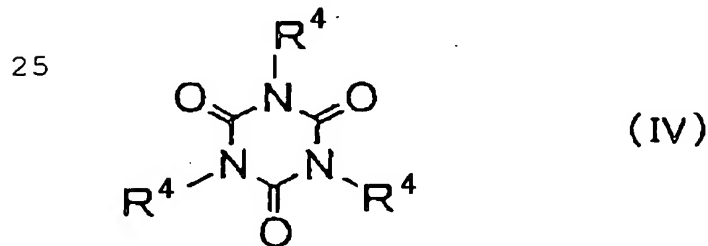
Preferred as the component (A), which may be mixed together to be used, from the viewpoint of less coloration, high
 10 optical transparency and high light resistance of the curing product obtained are vinylcyclohexene, dicyclopentadiene, triallyl isocyanurate, 2,2-bis(4-hydroxycyclohexyl)propane diallyl ether, and 1,2,4-trivinylcyclohexane. Particularly preferred are triallyl isocyanurate,
 15 2,2-bis(4-hydroxycyclohexyl)propane diallyl ether, and 1,2,4-trivinylcyclohexane.

[0042]

From the high heat resistance and transparency viewpoint, the component (A) is particularly preferably a
 20 triallylisocyanurate and derivatives thereof represented by the following general formula (IV):

[0043]

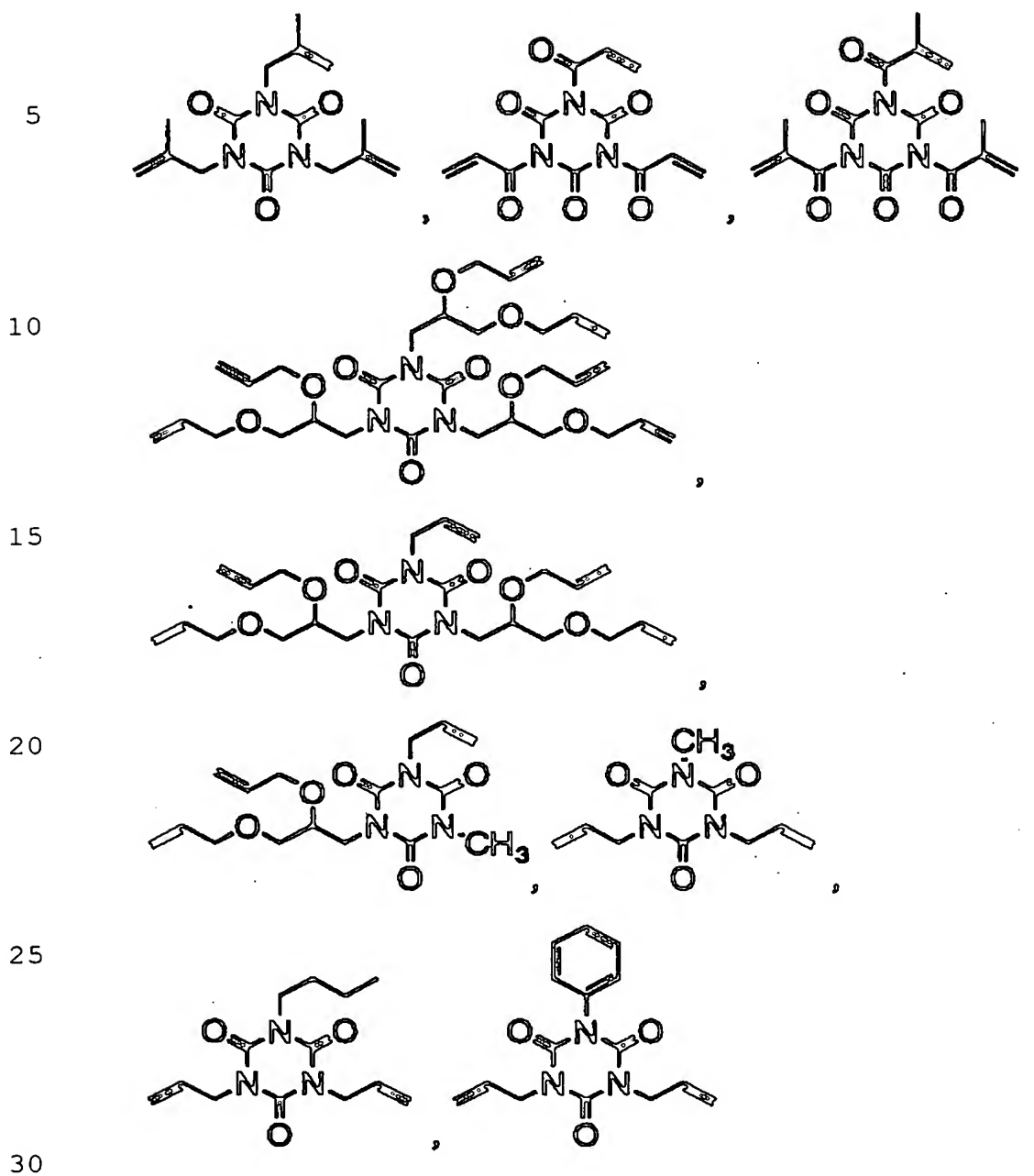
[Chemical 19]



30 wherein each R^4 represents a univalent organic group containing 1 to 50 carbon atoms and the three R^4 groups may be the same or different. As preferred specific examples of the organic compound represented by the general formula (IV), there may be mentioned triallyl isocyanurate,

35 [0044]

[Chemical 20]



and the like.

[0045]

An explanation is now made of the component (B), a compound containing a SiH group.

35 [0046]

The component (B) of the invention is a compound containing at least two SiH groups in each molecule.

[0047]

The component (B) may be any compound containing at least two SiH groups in each molecule, without any other restriction. Thus, for example, those compounds which are described in International Laid-open Patent Application WO 96/15194 and have at least two SiH groups in each molecule and the like can be used.

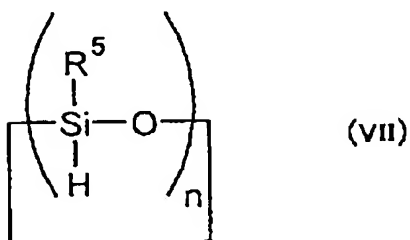
10 [0048]

From the availability viewpoint, linear and/or cyclic organopolysiloxanes having at least two SiH groups in each molecule are preferred, and, from the viewpoint of compatibility with the component (A), those cyclic organopolysiloxanes having at least two SiH groups in each molecule which are represented by the following general formula (III):

[0049]

[Chemical 21]

20



25

wherein R^5 represents an organic group containing 1 to 6 carbon atoms and n represents a number of 3 to 10, are more preferred.

[0050]

The substituent R^5 in the compounds represented by the general formula (VII) is preferably a substituent containing C, H and/or O, more preferably a hydrocarbon group, still more preferably a methyl group.

[0051]

From the ready availability viewpoint,
35 1,3,5,7-tetramethylcyclotetrasiloxane is preferred as the

compound represented by the general formula (VII).

[0052]

The molecular weight of the component (B) is not particularly restricted but may be optionally selected. From
5 the viewpoint that the fluidity can be manifested more easily, low molecular weights are preferred. In this case, the lower limit to the molecular weight is preferably 50, and the upper limit thereto is preferably 100,000, more preferably 1,000, still more preferably 700.

10 [0053]

The component (B) to be used may contain one single species or a mixture of two or more species.

[0054]

Further, from the viewpoint of good compatibility with
15 the component (A) and from the viewpoint that the component (B) is low in volatility and the problem of outgassing from the resulting composition hardly occurs, those compounds which can be obtained by subjecting an organic compound (α) having one or more carbon-carbon double bonds reactive with a SiH group
20 in each molecule and a linear and/or cyclic polyorganosiloxane (β) having at least two SiH groups in each molecule to hydrosilylation reaction are preferred as the component (B).

(Component (α))

25 Usable here as the component (α) are organic compounds (α 1) which have at least two carbon-carbon double bonds reactive with a SiH group in each molecule and are to serve as the component (A) mentioned above. The use of such component (α 1) tends to result in a high crosslink density and high mechanical
30 strength of the curing product.

[0055]

In addition, as the component (α), compounds (α 2) having one carbon-carbon double bond reactive with a SiH group in each molecule can also be used. The use of the component (α 2) tends
35 to render the curing product low in elasticity.

(Component ($\alpha 2$))

The component ($\alpha 2$) is not particularly restricted but may be any organic compound having one carbon-carbon double bond reactive with a SiH group in each molecule. From the viewpoint of better compatibility between the component (B) and the component (A), however, a compound other than a polysiloxane-organic block copolymer, a polysiloxane-organic graft copolymer or a like siloxane unit (Si-O-Si)-containing one is preferred, and a compound containing C, H, N, O, S and halogen, as exclusive constituent elements, is preferred.

[0056]

The sites of occurrence of the carbon-carbon double bonds reactive with a SiH group in component ($\alpha 2$) are not particularly restricted but may be anywhere within the molecule.

[0057]

The component ($\alpha 2$) compound may be classified as a polymer type compound or as a monomer type compound.

[0058]

Usable as the polymer type compound, there may be mentioned, for example, polysiloxane type, polyether type, polyester type, polyarylate type, polycarbonate type, saturated hydrocarbon type, unsaturated hydrocarbon type, polyacrylic ester type, polyamide type, phenol-formaldehyde type (phenol resin type), polyimide type compounds, etc.

[0059]

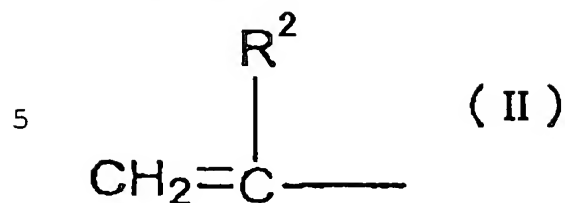
As the monomer type compound, there may be mentioned, for example, phenol type, bisphenol type, benzene, naphthalene or like aromatic hydrocarbon type compounds; aliphatic (e.g. chain, alicyclic, etc.) hydrocarbon type compounds; heterocyclic type compounds; silicon compounds; mixtures of these; etc.

[0060]

The carbon-carbon double bonds reactive with a SiH group in the component ($\alpha 2$) are not particularly restricted but, from the reactivity viewpoint, those groups represented by the following general formula (II):

[0061]

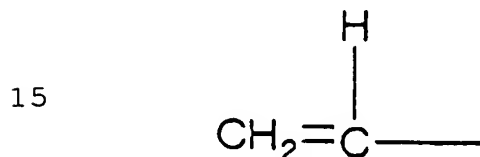
[Chemical 22]



wherein R^2 represents a hydrogen atom or a methyl group, are preferred. From the material availability viewpoint, the group represented by

[0062]

[Chemical 23]



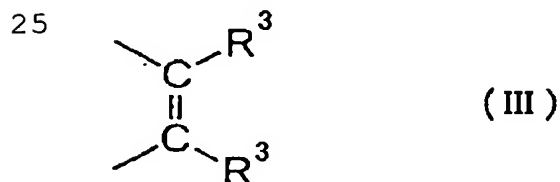
is particularly preferred.

[0063]

Furthermore, preferred as the carbon-carbon double bonds reactive with a SiH group in the component ($\alpha 2$) are alicyclic groups having a partial structure represented by the following general formula (III):

[0064]

[Chemical 24]

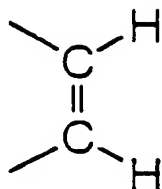


wherein each R^3 represents a hydrogen atom or a methyl group, in the ring structure, since high heat resistance can be given to curing products. From the material availability viewpoint, alicyclic groups having a partial structure represented by the following formula

35 [0065]

[Chemical 25]

5



in the ring structure are preferred.

The carbon-carbon double bonds reactive with a SiH group each may be directly bonded to the skeletal portion of the component (α^2) or covalently bonded thereto via a bivalent or polyvalent substituent. The above bivalent or polyvalent substituent is not particularly restricted but is preferably a substituent containing 0 to 10 carbon atoms. From the viewpoint of better compatibility between the component (B) and the component (A), however, a substituent containing C, H, N, O, S and halogen, as exclusive constituent elements is preferred. Examples of the above substituent are:

[0066]

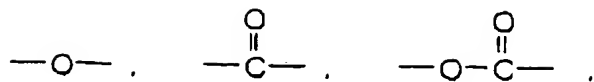
[Chemical 26]

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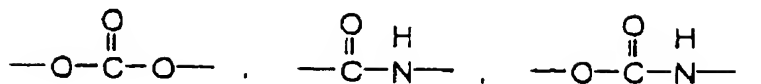
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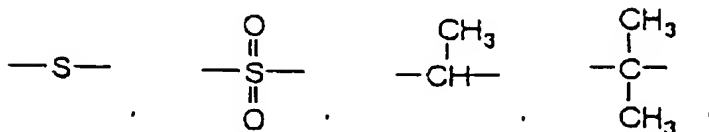
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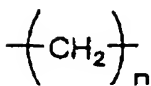
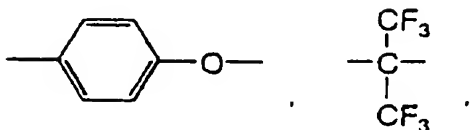
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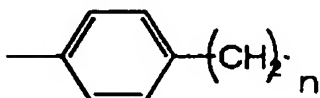
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15



20 (n representing a number of 1 to 10),



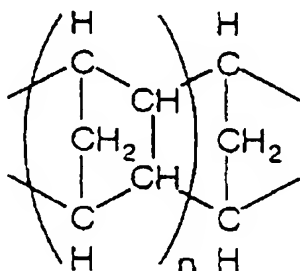
25

(n representing a number of 0 to 4), and

[0067]

[Chemical 27]

30



35

(n representing a number of 0 to 4), etc. Two or more of the bivalent or polyvalent substituents recited above may be

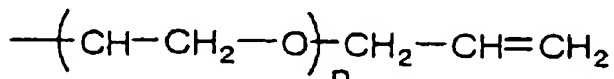
covalently bonded together to form one bivalent or polyvalent substituent.

[0068]

As examples of the group covalently bonded to the skeletal
 5 portion, there may be mentioned vinyl, allyl, methallyl, acryl,
 methacryl, 2-hydroxy-3-(allyloxy)propyl, 2-allylphenyl,
 3-allylphenyl, 4-allylphenyl, 2-(allyloxy)phenyl,
 3-(allyloxy)phenyl, 4-(allyloxy)phenyl, 2-(allyloxy)ethyl,
 2,2-bis(allyloxymethyl)butyl,
 10 3-allyloxy-2,2-bis(allyloxymethyl)propyl,

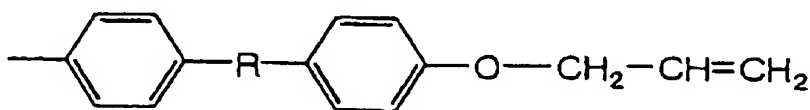
[0069]

[Chemical 28]



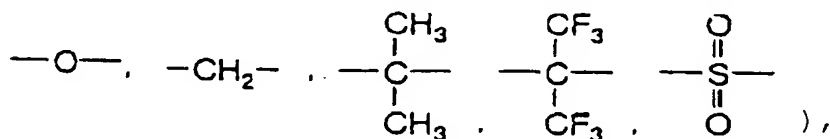
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(n representing a number satisfying the relation $5 \geq n \geq 2$),

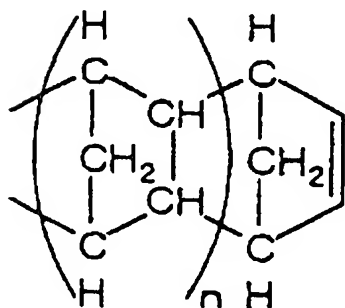


20

(R representing a bivalent group selected from among



25



30

(n representing a number of 0 to 4), and the like.

[0070]

35

As specific examples of the component ($\alpha 2$), there may be

mentioned linear aliphatic hydrocarbon compounds such as propene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 1-dodecene, 1-undecene, Idemitsu Petrochemical's Linealene, 4,4-dimethyl-1-pentene, 5 2-methyl-1-hexene, 2,3,3-trimethyl-1-butene and 2,4,4-trimethyl-1-pentene; alicyclic hydrocarbon compounds such as cyclohexene, methylcyclohexene, methylenecyclohexane, norbornylene, ethylidenecyclohexane, vinylcyclohexane, camphene, carene, α -pinene and β -pinene; aromatic hydrocarbon 10 compounds such as styrene, α -methylstyrene, indene, phenylacetylene, 4-ethynyltoluene, allylbenzene and 4-phenyl-1-butene; allyl ethers such as alkyl allyl ethers and allyl phenyl ether; aliphatic compounds such as glycerol monoallyl ether, ethylene glycol monoallyl ether and 15 4-vinyl-1,3-dioxolan-2-one; aromatic compounds such as 1,2-dimethoxy-4-allylbenzene and o-allylphenol; substituted isocyanurates such as monoallyl dibenzyl isocyanurate and monoallyl diglycidyl isocyanurate; silicon compounds such as vinyltrimethylsilane, vinyltrimethoxysilane and 20 vinyltriphenylsilane; etc.

Furthermore, as the component (α_2), there may be mentioned polymers and oligomers having a vinyl group at one end, for example polyether resins such as polyethylene oxide allylated at one end and polypropylene oxide allylated at one 25 end; hydrocarbon resins such as polyisobutylene allylated at one end; acrylic resins such as poly(butyl acrylate) allylated at one end and poly(methyl methacrylate) allylated at one end; etc.

[0071]

30 The structure may be linear or branched. The molecular weight is not particularly restricted but may be at any of various levels. The molecular weight distribution is not particularly restricted, either, but may be diverse. From the viewpoint that the mixture viscosity tends to become low and 35 the moldability tends to become better, however, the molecular

weight distribution is preferably not more than 3, more preferably not more than 2, still more preferably not more than 1.5.

[0072]

5 The glass transition temperature of the component (α_2), if any, is not particularly restricted, either, but may be diverse. In view of the tendency for the curing product obtained to become tough, however, the glass transition temperature is preferably not higher than 100°C, more
10 preferably not higher than 50°C, still more preferably not higher than 0°C. As preferred examples of the resin, there may be mentioned poly(butyl acrylate) and the like. Conversely, in view of the tendency for the curing product obtained to become highly resistant to heat, the glass transition temperature is
15 preferably not lower than 100°C, more preferably not lower than 120°C, still more preferably not lower than 150°C, most preferably not lower than 170°C.

 The glass transition temperature can be determined by dynamic viscoelasticity measurement and expressed in terms of
20 the temperature at which a maximum $\tan\delta$ value is obtained.

[0073]

 In view of the tendency for the curing product obtained to become high in heat resistance, the component (α_2) is preferably a hydrocarbon compound. In this case, the lower
25 limit to the number of carbon atoms is 7, and a preferred upper limit thereto is 10.

[0074]

 The component (α_2) may have other reactive group. As the above reactive group, there may be mentioned the following
30 groups, for example: epoxy, amino, radical-polymerizable unsaturated group, carboxyl, isocyanate, hydroxyl, alkoxysilyl group and so on. In cases where the above component has the above functional group, the adhesive properties of the curable composition obtained tends to become higher and the
35 strength of the curing product obtained tends to become high.

From the higher adhesive properties viewpoint, an epoxy group is preferred among those functional groups. In view of the tendency for the resulting curing product to have a higher level of heat resistance, it is preferred that the above component
5 have not less than one reactive group mentioned above per molecule on average. As specific examples, there may be mentioned monoallyl diglycidyl isocyanurate, allyl glycidyl ether, allyloxyethyl methacrylate, allyloxyethyl acrylate, vinyltrimethoxysilane, etc.

10 [0075]

The components (α 2) may be used singly or in combination of a plurality thereof.

(Component (β))

15 The component (β) is a linear and/or cyclic polyorganosiloxane having at least two SiH groups in each molecule.

[0076]

Specifically, there may be mentioned:

20 [0077]

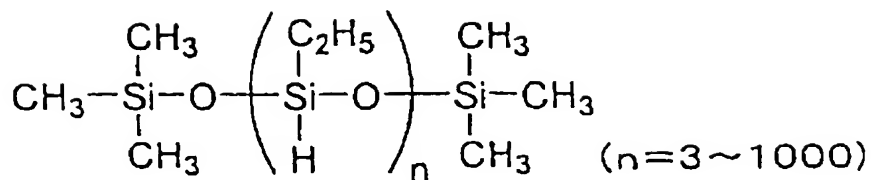
[Chemical 29]

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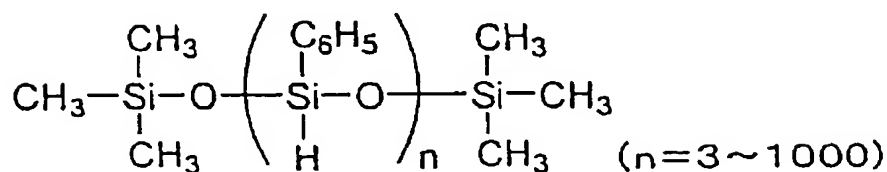
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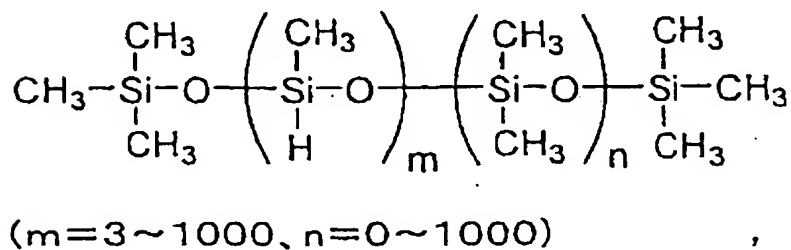
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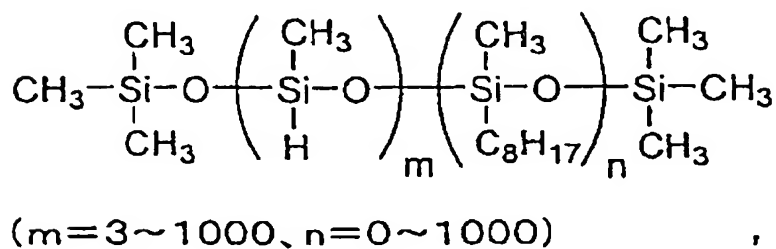
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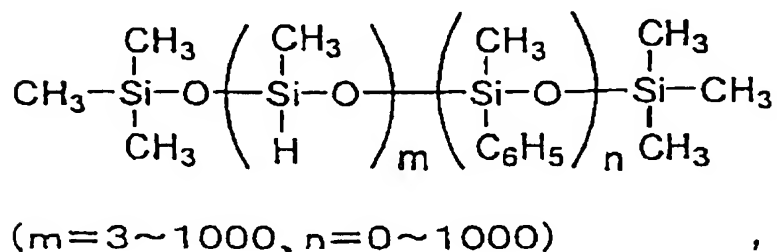
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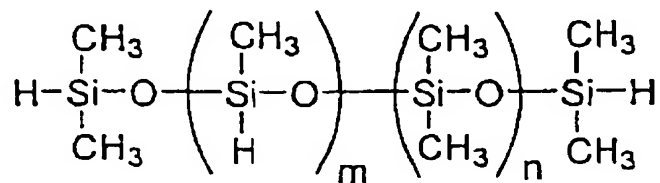
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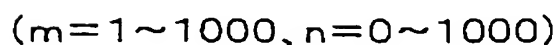
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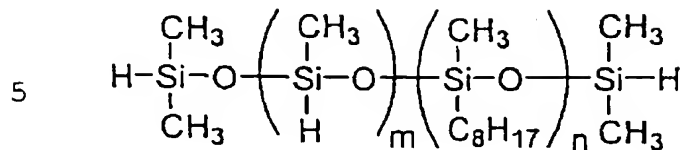
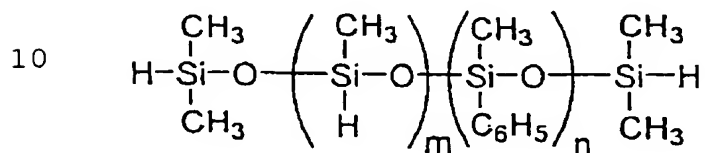
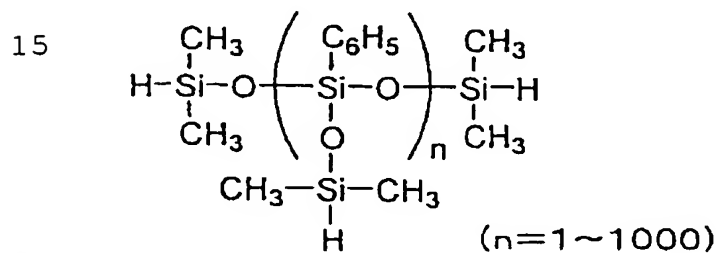
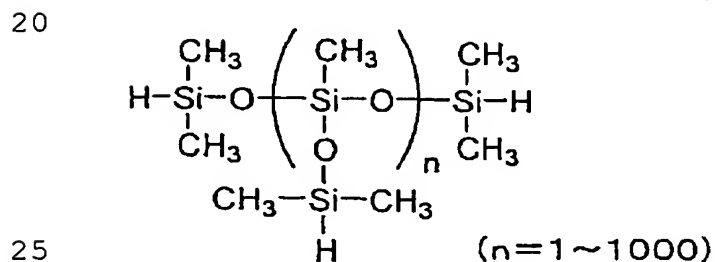
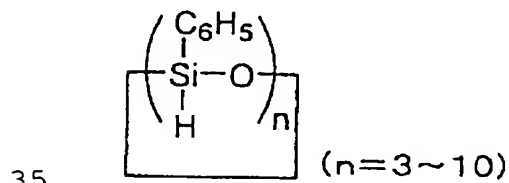
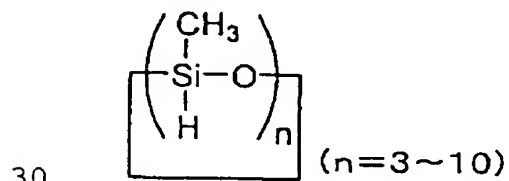


35



[0078]

[Chemical 30]

 $(m=1\sim1000, n=0\sim1000)$  $(m=1\sim1000, n=0\sim1000)$  $(n=1\sim1000)$  $(n=1\sim1000)$ 

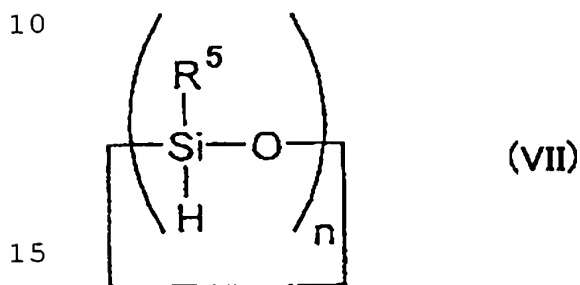
and the like.

[0079]

From the viewpoint that the compatibility with the component (α) tends to become better, those cyclic polyorganosiloxanes having at least three SiH groups in each molecule which are represented by the following general formula (VII):

[0080]

[Chemical 31]



wherein R^5 represents an organic group containing 1 to 6 carbon atoms and n represents a number of 3 to 10, are preferred.

[0081]

The substituent R^5 in the compounds represented by the general formula (VII) is preferably a substituent comprising C, H and/or O, more preferably a hydrocarbon group, still more preferably a methyl group.

[0082]

As the compounds represented by the general formula (IX), 1,3,5,7-tetramethylcyclotetrasiloxane is preferred from the ready availability viewpoint, etc.

[0083]

The component (β) may be used singly or as a mixture of two or more of them.

(Reaction between component (α) and component (β))

An explanation is now made of the hydrosilylation reaction between the component (α) and component (β), which is to be carried out in preparing a compound resulting from

hydrosilylation reaction between the component (α) and component (β) for use as the component (B) of the invention.

[0084]

The hydrosilylation reaction of the component (α) and component (β) may give a mixture of a plurality of compounds, including the component (B) of the invention in some instances. It is also possible to use the mixture as such in preparing the curable composition of the invention, without separating the component (B).

10 [0085]

The mixing ratio between the component (α) and component (β) in carrying out the hydrosilylation reaction between the component (α) and component (β) is not particularly restricted but, when the strength of the curing product resulting from hydrosilylation of the component (A) with the component (B) obtained is taken into consideration, a higher SiH group content in the component (B) is preferred, so that the ratio of the total number (X) of carbon-carbon double bonds reactive with a SiH group as occurring in the component (α) to be admixed and the total number (Y) of SiH groups in the component (β) to be admixed is preferably such that it is not less than 2 ($Y/X \geq 2$), more preferably such that it is not less than 3 ($Y/X \geq 3$). From the viewpoint that the compatibility of the component (B) with the component (A) tends to become better, it is preferred that the ratio satisfied the relation $10 \geq Y/X$, more preferably $5 \geq Y/X$.

[0086]

In subjecting the component (α) and component (β) to hydrosilylation reaction, an appropriate catalyst may be used. As the catalyst, there may be mentioned simple substance platinum, solid platinum supported on such a carrier as alumina, silica or carbon black, chloroplatinic acid, complexes of chloroplatinic acid with an alcohol, aldehyde, ketone, etc., platinum-olefin complexes (e.g. $\text{Pt}(\text{CH}_2=\text{CH}_2)_2(\text{PPh}_3)_2$, $\text{Pt}(\text{CH}_2=\text{CH}_2)_2\text{Cl}_2$), platinum-vinylsiloxane complexes (e.g. $\text{Pt}(\text{ViMe}_2\text{SiOSiMe}_2\text{Vi})_n$, $\text{Pt}[(\text{MeViSiO})_4]_m$), platinum-phosphine

complexes (e.g. $\text{Pt}(\text{PPh}_3)_4$, $\text{Pt}(\text{PBu}_3)_4$), platinum-phosphite complexes (e.g. $\text{Pt}[\text{P}(\text{OPh})_3]_4$, $\text{Pt}[\text{P}(\text{OBu})_3]_4$) (in the formulas, Me represents a methyl group, Bu a butyl group, Vi a vinyl group and Ph a phenyl group, and n and m each represents an integer),
 5 dicarbonyldichloroplatinum, Karstedt catalyst, platinum-hydrocarbon complexes described in Ashby's U. S. Patent No. 3,159,601 and 3,159,662, platinum-alcoholate catalysts described in Lamoreaux's U. S. Patent No. 3,220,972, and so on. Furthermore, platinum chloride-olefin complexes
 10 described in Modic's U. S. Patent No. 3,516,946 are also useful in the practice of the present invention.

[0087]

As catalysts other than platinum compounds, there may be mentioned, for example, $\text{RhCl}(\text{PPh})_3$, RhCl_3 , RhAl_2O_3 , RuCl_3 , IrCl_3 ,
 15 FeCl_3 , AlCl_3 , $\text{PdCl}_2 \cdot 2\text{H}_2\text{O}$, NiCl_2 , TiCl_4 , etc.

[0088]

Among these, chloroplatinic acid, platinum-olefin complexes, platinum-vinylsiloxane complexes and the like are preferred from the catalytic activity viewpoint. These
 20 catalysts may be used singly or in combination of two or more.

[0089]

The level of addition of the catalyst is not particularly restricted but, for attaining a sufficient level of curability and reducing the cost of the curable composition to a relatively
 25 low level, the lower limit to the addition level is preferably 10^{-8} mole, more preferably 10^{-6} mole, per mole of the SiH group in the component (β), and the upper limit to the addition level is preferably 10^{-1} mole, more preferably 10^{-2} mole, per mole of the SiH group in the component (β).

30 [0090]

A promoter may be used in combination with the above catalyst. As the promoter, there may be mentioned, for example, phosphorus compounds such as triphenylphosphine; 1,2-diester compounds such as dimethyl maleate; acetylene alcohol compounds
 35 such as 2-hydroxy-2-methyl-1-butyne; sulfur compounds

inclusive of simple substance sulfur; amine compounds such as triethylamine; etc. The level of addition of the promoter is not particularly restricted but the lower limit to the addition level is preferably 10^{-2} mole, more preferably 10^{-1} mole, and
5 the upper limit is 10^2 mole, more preferably 10 moles, per mole of said hydrosilyl catalyst.

[0091]

As for the method of mixing up the component (α), component (β) and catalyst in carrying out the reaction, various
10 methods may be employed. Preferred is, however, the method comprising mixing the component (α) with the catalyst and mixing the resulting mixture with the component (β). The method comprising mixing the catalyst with a mixture of the component (α) and component (β) makes it difficult to control the reaction.
15 When the method comprising mixing the component (α) with a mixture of the component (β) and catalyst is employed, the component (β), which is reactive with moisture possibly contained therein in the presence of the catalyst, may cause denaturation in some instances.

20 [0092]

The reaction temperature may be selected at various levels. In the instance case, the lower limit to the reaction temperature is preferably 30°C , more preferably 50°C , and the upper limit to the reaction temperature is preferably 200°C ,
25 more preferably 150°C . At lower reaction temperatures, the reaction time for the reaction to proceed to a satisfactory extent becomes long whereas higher reaction temperatures are not practical. The reaction may be carried out at a constant temperature, or the temperature may be varied stepwise or
30 continuously according to need.

[0093]

The reaction time and the pressure during reaction may also be selected at various levels according to need.

[0094]

35 A solvent may be used in the hydrosilylation reaction.

The solvent to be used is not particularly restricted but any of those which will not inhibit the hydrosilylation reaction. Specifically, hydrocarbon solvents such as benzene, toluene, hexane and heptane; ether solvents such as tetrahydrofuran, 1,4-dioxane, 1,3-dioxolane and diethyl ether; ketone solvents such as acetone and methyl ethyl ketone; and halogenated solvents such as chloroform, methylene chloride and 1,2-dichloroethane can be used satisfactorily. Such solvents can also be used in the form of a mixed solvent composed of two or more species. Preferred as the solvent are toluene, tetrahydrofuran, 1,3-dioxolane and chloroform. The amount of the solvent to be used can be adequately selected.

[0095]

Further, various additives may be used for the purpose of controlling the reactivity, etc.

[0096]

It is also possible, after reacting the component (α) with the component (β), to remove the solvent, unreacted component (α) and/or unreacted component (β). When these volatile components are removed, the component (B) obtained becomes free of such volatile components and, therefore, the problems of void formation and cracking due to volatile matter evaporation in the step of curing with the component (A) will hardly occur. The method of removal includes, for example, volatile matter removal under reduced pressure and, further, treatment with active carbon, aluminum silicate, silica gel or the like. In the case of volatile matter removal under reduced pressure, the treatment is preferably carried out at low temperatures. In this case, the upper limit to the temperature is preferably 100°C, more preferably 60°C. High temperature treatment tends to be accompanied by changes in properties, for example viscosity increase.

[0097]

As examples of the component (B), namely a reaction product from the components (α) and (β), there may be mentioned

the bisphenol A diallyl
ether-1,3,5,7-tetramethylcyclotetrasiloxane reaction product,
vinylcyclohexene-1,3,5,7-tetramethylcyclotetrasiloxane
reaction product,
5 divinylbenzene-1,3,5,7-tetramethylcyclotetrasiloxane
reaction product,
dicyclopentadiene-1,3,5,7-tetramethylcyclotetrasiloxane
reaction product, triallyl
isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction
10 product, diallyl monoglycidyl
isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction
product, allyl glycidyl
ether-1,3,5,7-tetramethylcyclotetrasiloxane reaction product,
(α)-methylstyrene-1,3,5,7-tetramethylcyclotetrasiloxane
15 reaction product, monoallyl diglycidyl
isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction
product, etc. From the viewpoint of the heat resistance and
light resistance, the triallyl
isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction
20 product, diallyl monoglycidyl
isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction
product, and monoallyl diglycidyl
isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction
product are preferred.

25 [0098]

The component (C), a hydrosilylation catalyst is now
explained.

[0099]

The hydrosilylation catalyst is not particularly
30 restricted but may be any one having catalytic activity in the
hydrosilylation reaction. Thus, there may be mentioned, for
example, simple substance platinum, solid platinum supported
on such a carrier as alumina, silica or carbon black,
chloroplatinic acid, complexes of chloroplatinic acid with an
35 alcohol, aldehyde, ketone, etc., platinum-olefin complexes

(e.g. $\text{Pt}(\text{CH}_2=\text{CH}_2)_2(\text{PPh}_3)_2$, $\text{Pt}(\text{CH}_2=\text{CH}_2)_2\text{Cl}_2$), platinum-vinylsiloxane complexes (e.g. $\text{Pt}(\text{ViMe}_2\text{SiOSiMe}_2\text{Vi})_n$, $\text{Pt}[(\text{MeViSiO})_4]_m$), platinum-phosphine complexes (e.g. $\text{Pt}(\text{PPh}_3)_4$, $\text{Pt}(\text{PBu}_3)_4$), platinum-phosphite complexes (e.g. $\text{Pt}[\text{P}(\text{OPh})_3]_4$,
 5 $\text{Pt}[\text{P}(\text{OBu})_3]_4$) (in the formulas, Me represents a methyl group, Bu a butyl group, Vi a vinyl group and Ph a phenyl group, and n and m each represents an integer), dicarbonyldichloroplatinum, Karstedt catalyst, platinum-hydrocarbon complexes described in Ashby's U. S. Patents No. 3,159,601 and 3,159,662,
 10 platinum-alcoholate catalysts described in Lamoreaux's U. S. Patent No. 3,220,972, and so on. Furthermore, platinum chloride-olefin complexes described in Modic's U. S. Patent No. 3,516,946 are also useful in the practice of the present invention.

15 [0100]

As catalysts other than platinum compounds, there may be mentioned, for example, $\text{RhCl}(\text{PPh})_3$, RhCl_3 , RhAl_2O_3 , RuCl_3 , IrCl_3 , FeCl_3 , AlCl_3 , $\text{PdCl}_2 \cdot 2\text{H}_2\text{O}$, NiCl_2 , TiCl_4 , etc.

[0101]

20 Among these, chloroplatinic acid, platinum-olefin complexes, platinum-vinylsiloxane complexes and the like are preferred from the catalytic activity viewpoint. The above catalysts may be used singly or in combination of two or more.

[0102]

25 The level of addition of the catalyst is not particularly restricted but, for attaining a sufficient level of curability and reducing the cost of the curable composition to a relatively low level, the lower limit to the addition level is preferably 10^{-8} mole, more preferably 10^{-6} mole, per mole of the SiH group
 30 in the component (B), and the upper limit to the addition level is preferably 10^{-1} mole, more preferably 10^{-2} mole, per mole of the SiH group in the component (B).

[0103]

A promoter may be used in combination with the above
 35 catalyst. As the promoter, there may be mentioned, for example,

phosphorus compounds such as triphenylphosphine; 1,2-diester compounds such as dimethyl maleate; acetylene alcohol compounds such as 2-hydroxy-2-methyl-1-butyne; sulfur compounds inclusive of simple substance sulfur; amine compounds such as triethylamine; etc. The level of addition of the promoter is not particularly restricted but the lower limit to the addition level is preferably 10^{-2} mole, more preferably 10^{-1} mole, and the upper limit is 10^2 mole, more preferably 10 moles, per mole of said hydrosilylation catalyst.

10 [0104]

A curing retarder may be incorporated for improving the storage stability of the composition of the invention or for adjusting the reactivity in the hydrosilylation reaction in the course of production. As the curing retarder, there may be mentioned aliphatic unsaturated bond-containing compounds, organophosphorus compounds, organosulfur-containing compounds, nitrogen-containing compounds, tin compounds, organic peroxides, etc., and two or more of these may be used in combination.

20 The aliphatic unsaturated bond-containing compounds include, for example, propargyl alcohols, en-yne compounds, maleic acid esters, etc. The organophosphorus compounds include, for example, triorganophosphines, diorganophosphines, organophosphones, triorganophosphites, etc. The organosulfur-containing compounds include, organomercaptans, diorganosulfides, hydrogen sulfide, benzothiazole, benzothiazole disulfide, etc. The nitrogen-containing compounds include ammonia, primary to tertiary alkylamines, arylamines, urea, hydrazine, etc. The tin compounds include stannous halide dehydrates, stannous carboxylates, etc. The organic peroxides include di-tert-butyl peroxide, dicumyl peroxide, benzoyl peroxide, tert-butyl perbenzoate, etc.

[0105]

From the good retarder activity and availability viewpoint, benzothiazole, thiazole, dimethyl maleate, and

35

2-hydroxy-2-methyl-3-butyne are preferred among the above curing retarders.

[0106]

5 The level of addition of the above curing retarder can be selected at various levels but the lower limit to the addition level is preferably 10^{-1} mole, more preferably 1 mole, and the upper limit to the addition level is preferably 10^3 moles, more preferably 50 moles, per mole of the hydrosilylation catalyst.

[0107]

10 These curing retarders may be used singly or two or more of them may be used in combination.

[0108]

15 To the composition of the invention, an adhesion property-improving agent may be incorporated. The adhesion property-improving agent includes, for example, adhesives in general use, a coupling agent, epoxy resin, phenol resin, coumarone-indene resin, rosin ester resin, terpene-phenol resin, α -methylstyrene-vinyl toluene copolymer, polyethyl methylstyrene, aromatic polyisocyanate, and the like.

20 [0109]

As examples of the above coupling agent, there may be mentioned a silane coupling agent. The silane coupling agent is not particularly restricted but may be any of those compounds which have, within the molecule, at least one functional group reactive with an organic group and at least one hydrolyzable silyl group. The functional group reactive with an organic group is preferably at least one functional group selected from among epoxy, methacryl, acryl, isocyanate, isocyanurate, vinyl and carbamate groups from the handleability viewpoint. From
25 the viewpoint of curability and adhesive properties, epoxy, methacryl and acryl groups are particularly preferred. The hydrolyzable silyl group is preferably an alkoxysilyl group and, from the reactivity viewpoint, a methoxysilyl and an
30 ethoxysilyl groups are particularly preferred.

35 [0110]

As preferred examples of the silane coupling agent, there may be mentioned epoxy functional group-containing alkoxysilanes such as 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropyltriethoxysilane,
 5 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane and 2-(3,4-epoxycyclohexyl)ethyltriethoxysilane; and methacryl or acryl group-containing alkoxysilanes such as 3-methacryloxypropyltrimethoxysilane, 3-methacryloxypropyltriethoxysilane,
 10 3-acryloxypropyltrimethoxysilane, 3-acryloxypropyltriethoxysilane, methacryloxymethyltrimethoxysilane, methacryloxymethyltriethoxysilane, acryloxymethyltrimethoxysilane and
 15 acryloxymethyltriethoxysilane.

[0111]

The level of addition of the silane coupling agent can be selected at various levels. However, the lower limit to the addition level is preferably 0.1 part by weight, more preferably
 20 0.5 part by weight, and the upper limit to the addition level is preferably 50 parts by weight, more preferably 25 parts by weight, per 100 parts by weight of [component (A) + component (B)]. At lower addition levels, the adhesive property-improving effect is hardly exerted and, at higher
 25 addition levels, the physical properties of the curing product may possibly be adversely affected.

[0112]

The epoxy group-containing compound includes, for example, novolak phenol-based epoxy resins, biphenyl-based
 30 epoxy resins, dicyclopentadiene-based epoxy resins, bisphenol F diglycidyl ether, bisphenol A diglycidyl ether, 2,2'-bis(4-glycidylloxycyclohexyl)propane, 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexanecarboxylate, vinylcyclohexene dioxide,
 35 2-(3,4-epoxycyclohexyl)-5,5-spiro(3,4-epoxycyclohexane)-1,3

-dioxane, bis(3,4-epoxycyclohexyl) adipate, bisglycidyl 1,2-cyclopropanedicarboxylate, triglycidyl isocyanurate, monoallyl diglycidyl isocyanurate, diallyl monoglycidyl isocyanurate, and the like.

5 [0113]

The level of addition of the epoxy group-containing compound can be selected at various levels. However, the lower limit to the addition level is preferably 1 part by weight, more preferably 3 part by weight, and the upper limit to the addition level is preferably 50 parts by weight, more preferably 25 parts by weight, per 100 parts by weight of [component (A) + component (B)]. At lower addition levels, the adhesive property-improving effect is hardly exerted and, at higher addition levels, the physical properties of the curing product may possibly be adversely affected.

[0114]

The coupling agents, silane coupling agents, and epoxy group-containing compounds etc. may be used each singly or two or more of them may be used in combination.

20 [0115]

In the present invention, a silanol condensation catalyst may further be incorporated for improving the effect of the coupling agents and epoxy group-containing compounds such that improvement of the adhesive properties and/or stabilization are possible. The silanol condensation catalyst is not particularly restricted, but aluminum compounds and/or titanium compounds and/or borate esters are preferred.

As the above aluminum compound to be a silanol condensation catalyst, for example, there may be mentioned aluminum alkoxides such as triisopropoxyaluminum, sec-butoxy aluminum diisopropoxide, and aluminum tri-sec-butoxide; and aluminum chelate compounds such as aluminum ethyl acetoacetate diisopropoxide, aluminum tris(ethyl acetoacetate), Alumi-chelate M (product of Kawaken Fine Chemicals Co., Ltd., alkyl acetoacetate aluminum diisopropoxide), aluminum

tris(acetylacetonate), and aluminum bis(ethyl acetoacetate) monoacetylacetonate; etc. From the viewpoint of handling, aluminum chelate compounds are preferred. As the titanium compounds mentioned above to be silanol condensation catalyst, for example, there may be mentioned tetraalkoxytitaniums such as tetraisopropoxytitanium and tetrabutoxytitanium; titanium chelates such as titanium tetraacetylacetonate; common titanate coupling agents having such a residue as oxyacetic acid or ethylene glycol; etc.

10 [0116]

The borate esters are used as silanol condensation catalysts such that improvement of the adhesive properties and/or stabilization are possible. As the borate esters, a compound represented by the following general formula (VIII) or (VIII'):

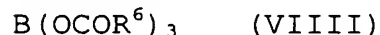
15 [0117]

[Chemical 32]



[0118]

20 [Chemical 33]



As specific borate esters, tri-2-ethylhexyl borate, normaltrioctadecyl borate, trinormaloctyl borate, triphenyl borate, trimethylene borate, tris(trimethylsilyl) borate, trinormalbutyl borate, tri-sec-butyl borate, tri-tert-butyl borate, triisopropyl borate, trinormalpropyl borate, triallyl borate, triethyl borate, trimethyl borate, boron methoxyethoxide and the like may be suitably used. The borate esters may be used singly or two or more of them may be used in combination. The mixing may be carried out preliminary, or during cured product-production.

[0119]

35 From the availability viewpoint, trimethyl borate,

triethyl borate, and trinormalbutyl borate are more preferred, and trimethyl borate is still more preferred.

[0120]

From the viewpoint of the possibility of volatility suppression in the step of curing, trinormaloctadecyl borate, 5 trinormaloctyl borate, triphenyl borate, trimethylene borate, tris(trimethylsilyl) borate, trinormalbutyl borate, tri-sec-butyl borate, tri-tert-butyl borate, triisopropyl borate, trinormalpropyl borate, triallyl borate, and boron 10 methoxyethoxide are preferred, and normaltrioctadecyl borate, tri-tert-butyl borate, triphenyl borate, and trinormalbutyl borate are still more preferred.

[0121]

From the volatility suppression and/or workability 15 viewpoint, trinormalbutyl borate, triisopropyl borate, and trinormalpropyl borate are more preferred, and trinormalbutyl borate is still more preferred.

From the viewpoint of low degrees of coloration at high temperatures, triethyl borate is more preferred, and trimethyl 20 borate is still more preferred.

[0122]

The level of addition of the silanol condensation catalyst can be selected at various levels. However, the lower limit to the addition level is preferably 0.1 part by weight, 25 more preferably 1 part by weight, and the upper limit to the addition level is preferably 50 parts by weight, more preferably 30 parts by weight, per 100 parts by weight of the coupling agent and/or epoxy-containing compound. At lower addition levels, the adhesive property-improving effect is hardly exerted and, 30 at higher addition levels, the physical properties of the curing product may possibly be adversely affected.

[0123]

The silanol condensation catalyst may be used singly or two or more of them may be used in combination.

35 [0124]

In the present invention, a silanol source compound may be used such that adhesive-property improving effect can be more enhanced, whereby further improvement of the adhesive properties and/or stabilization can be achieved. As the above
5 silanol source compound, there may be mentioned, for example, silanol compounds such as triphenyl silanol and diphenyldihydroxysilane, alkoxysilanes such as diphenyldimethoxysilane, tetramethoxysilane and methyltrimethoxysilane, etc.

10 [0125]

When such a silanol source compound is used, the amount thereof to be used can be selected at various levels. However, the lower limit to the addition level is preferably 0.1 part by weight, more preferably 1 part by weight, and the upper limit
15 to the addition level is preferably 50 parts by weight, more preferably 30 parts by weight, per 100 parts by weight of the coupling agent and/or the epoxy compound.

At lower addition levels, the adhesive property-improving effect is hardly exerted and, at higher
20 addition levels, the physical properties of the curing product may possibly be adversely affected.

[0126]

The above silanol source compounds may be used singly or two or more of them may be used in combination.

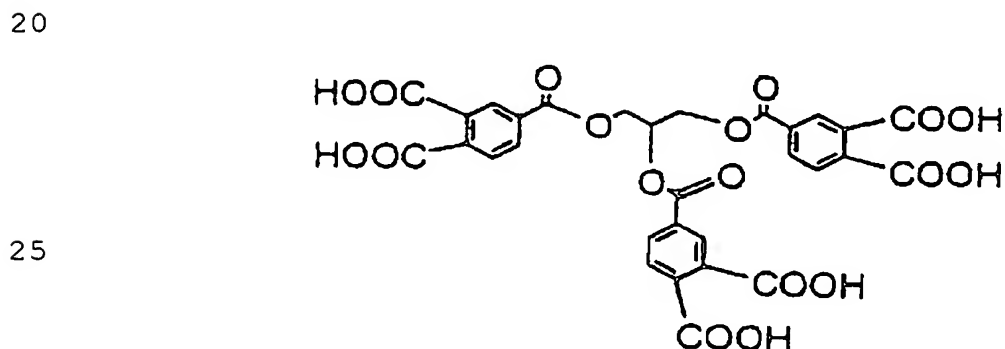
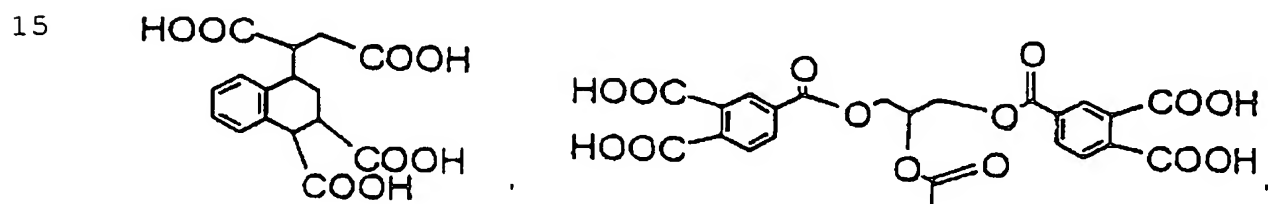
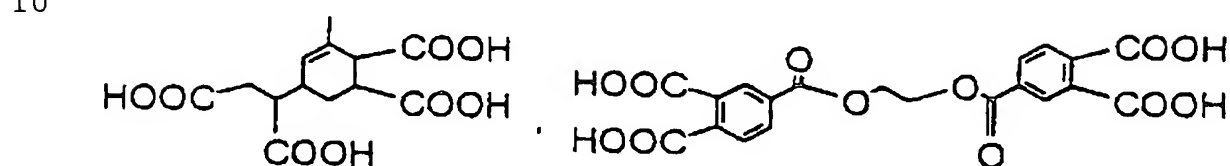
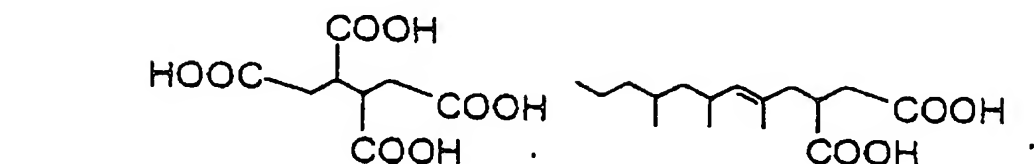
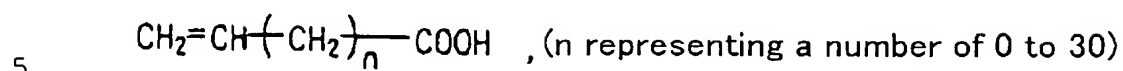
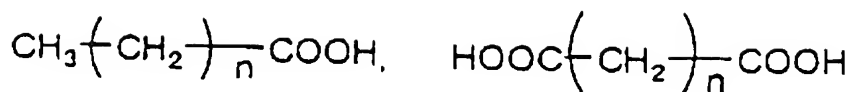
25 [0127]

In the present invention, carboxylic acids and/or acid anhydrides may be incorporated for improving the effect of the coupling agents and epoxy group-containing compounds such that improvement of the adhesive properties and/or stabilization are
30 possible. The above carboxylic acid or acid anhydride is not particularly restricted, but for example, includes the following:

[0128]

[Chemical 34]

35



30 2-ethylhexanoic acid, cyclohexanecarboxylic acid,
cyclohexanedicarboxylic acid, methylcyclohexanedicarboxylic
acid, tetrahydrophthalic acid, methyltetrahydrophthalic acid,
methylhimic acid, norbornenedicarboxylic acid, hydrogenated
methylnadac acid, maleic acid, acetylenedicarboxylic acid,
lactic acid, malic acid, citric acid, tartaric acid, benzoic
35 acid, hydroxybenzoic acid, cinnamic acid, phthalic acid,

trimellitic acid, pyromellitic acid, naphthalenecarboxylic acid, naphthalenedicarboxylic acid, and the like, as well as the acid anhydrides and mixed acid anhydrides thereof.

[0129]

5 Among the above carboxylic acids and acid anhydrides, those having a carbon-carbon double bond reactive with a SiH group are preferred since they have hydrosilylation reactivity, are less capable of exuding from the curing product and hardly impair the physical properties of the resulting curing product.
10 As preferred carboxylic acids and/or acid anhydrides, there may be mentioned, for example,

[0130]

[Chemical 35]

15
$$\text{CH}_2=\text{CH}\left(\text{CH}_2\right)_n\text{COOH} \quad (\text{n representing a number of 0 to 30}),$$

tetrahydrophthalic acid, methyltetrahydrophthalic acid, and anhydrides and mixed anhydrides thereof.

20 [0131]

When such a carboxylic acid and/or acid anhydride is used, the level of addition thereof can be selected at various levels. However, the lower limit to the addition level is preferably 0.1 part by weight, more preferably 1 part by weight, and the
25 upper limit to the addition level is preferably 50 parts by weight, more preferably 10 parts by weight, per 100 parts by weight of the coupling agent and/or epoxy-containing compound. At lower addition levels, the adhesive property-improving effect is hardly exerted and, at higher addition levels, the
30 physical properties of the curing product may possibly be adversely affected.

[0132]

The above carboxylic acids and/or acid anhydrides may be used singly or two or more of them may be used in combination.

35 [0133]

For modifying the characteristics of the curable composition of the invention, it is also possible to add thermosetting resins thereto. The above thermosetting resin is not particularly restricted, but there may be mentioned, for example, epoxy resins, cyanate ester resins, phenol resins, polyimide resins, urethane resins, bismaleimide resins, etc. Among them, epoxy resins are preferred from the viewpoint that they are excellent in the practical properties such as adhesiveness.

10 [0134]

The epoxy resin includes, for example, novolak phenol-based epoxy resins, biphenyl-based epoxy resins, dicyclopentadiene-based epoxy resins, bisphenol F diglycidyl ether, bisphenol A diglycidyl ether, 15 2,2'-bis(4-glycidylloxycyclohexyl)propane, 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexanecarboxylate, vinylcyclohexene dioxide, 2-(3,4-epoxycyclohexyl)-5,5-spiro(3,4-epoxycyclohexane)-1,3-dioxane, bis(3,4-epoxycyclohexyl) adipate, bisglycidyl 20 1,2-cyclopropanedicarboxylate, triglycidyl isocyanurate, monoallyl diglycidyl isocyanurate, diallyl monoglycidyl isocyanurate, and like epoxy resins as cured with an aliphatic acid anhydride such as hexahydrophthalic anhydride, methylhexahydrophthalic anhydride, 25 trialkyltetrahydrophthalic anhydrides or hydrogenated methyl nadic anhydride, and the like. The epoxy resin or a curing agent may respectively be used singly or in the form of a combination of a plurality of species.

[0135]

30 The level of addition of the above thermosetting resin is not particularly restricted. However, the lower limit to the amount to be used is preferably 5% by weight, more preferably 10% by weight, and the upper limit to the amount to be used is preferably 50% by weight, more preferably 30% by weight, of the 35 total weight of the curable composition. At lower addition

levels, the desired effects, such as adhesive properties, can hardly be obtained and, at higher levels, the curing product tends to become brittle.

[0136]

- 5 The thermosetting resin may comprise one single species or a combination of a plurality of species.

[0137]

- 10 A raw resin and/or a cured resin of the thermosetting resin may be mixed with the component (A) and/or (B) by homogeneously dissolving therein, dispersing therein in the form of ground particles, dispersing therein in the form of a solution in a solvent, or the like. From the viewpoint that the resulting curing product readily becomes more transparent, the resin is preferably mixed with the component (A) and/or (B) by
- 15 homogeneously dissolving therein. In this case, too, the thermosetting resin may be directly dissolved in the component (A) and/or (B) or homogeneously mixed therewith using a solvent or the like, or the solvent may be removed thereafter to attain a homogeneous dispersion state and/or mixture state.

- 20 [0138]

- When the thermosetting resin is dispersed, the average particle diameter can be selected at various levels. Preferably, however, the lower limit to the average particle diameter is 10 nm, and the upper limit to the average particle
- 25 diameter is preferably 10 μm . The particle system may have a distribution and may be monodisperse or show a plurality of peak particle diameters. From the viewpoint that the curable composition tend to be low in viscosity and show better moldability, the coefficient of variation in particle diameter
- 30 is preferably not more than 10%.

[0139]

- For modifying the characteristics of the composition of the invention, it is also possible to incorporate thermoplastic resins thereto. The above thermoplastic resin is not
- 35 particularly restricted, but for example, there may be

mentioned acrylic resins, typically poly(methyl methacrylate)-based resins such as methyl methacrylate homopolymers, or random, block or graft copolymers of methyl methacrylate and another monomer, or other polymers (e.g. Optorez, product of Hitachi Chemical, etc.), and poly(butyl acrylate)-based resins such as butyl acrylate homopolymers, or random, block or graft polymers of butyl acrylate and another monomer, or other polymers; polycarbonate-type resins such as polycarbonate resins containing bisphenol A, 3,3,5-trimethylcyclohexylidenebisphenol or the like as a monomer structure (e.g. APEC, product of Teijin, etc.); cycloolefin resins such as resins resulting from homopolymerization or copolymerization of norbornene derivatives, vinyl monomers and so forth, resins resulting from ring-opening metathetic polymerization of norbornene derivatives, or hydrogenation products derived therefrom (e.g. APEL, product of Mitsui Chemical; ZEONOR and ZEONEX, products of Zeon Corporation; ARTON, product of JSR; etc.); olefin-maleimide-based resins such as ethylene-maleimide copolymers (e.g. TI-PAS, product of Tosoh, etc.); polyester resins such as polyesters resulting from polycondensation of a bisphenol such as bisphenol A or bis(4-(2-hydroxyethoxy)phenyl)fluorine and/or a diol such as diethylene glycol and a phthalic acid such as terephthalic acid or isophthalic acid and/or an aliphatic dicarboxylic acid (e.g. O-PET, product of Kanebo, etc.); polyethersulfone resins; polyarylate resins; poly(vinyl acetal) resins; polyethylene resins; polypropylene resins; polystyrene resins; polyamide resins; silicone resins, fluororesins; rubber-like resins such as natural rubbers and EPDM; etc.

[0140]

The above thermoplastic resin may have a carbon-carbon double bond(s) reactive with a SiH group and/or a SiH group(s) within the molecule. In view of the tendency for the resulting curing product to have an increased level of toughness, it is

preferred that it has at least one carbon-carbon double bond reactive with a SiH group and/or at least one SiH group per molecule on average.

[0141]

5 The above thermoplastic resin may have another or other crosslinking group or groups. As the above crosslinking groups, there may be mentioned epoxy, amino, radical-polymerizable unsaturated groups, carboxyl, isocyanate, hydroxyl, alkoxy-silyl groups, etc. In view of the tendency for the
10 resulting curing product to have an increased level of heat resistance, the occurrence of at least one crosslinking group per molecule on average is preferred.

[0142]

15 The molecular weight of the above thermoplastic resin is not particularly restricted but, from the viewpoint of better compatibility with the component (A) and/or (B), the number average molecular weight is preferably not more than 10,000, more preferably not more than 5,000. Conversely, from the viewpoint of the tendency for the resulting curing product to
20 readily become tougher, the number average molecular weight is preferably not less than 10,000, more preferably not less than 100,000. The molecular weight distribution is not particularly restricted, either. In view of the tendency for the mixture to have a lower viscosity and better moldability,
25 however, the molecular weight distribution is preferably not more than 3, more preferably not more than 2, still more preferably not more than 1.5.

[0143]

30 The level of incorporation of the thermoplastic resin is not particularly restricted but the lower limit to the amount to be used is preferably 5% by weight, more preferably 10% by weight, of the whole amount of the curable composition, and the upper limit to the amount to be used is preferably 50% by weight, more preferably 30% by weight, in the curable composition. When
35 the addition level is lower, the curing product obtained tends

to be brittle while higher addition levels tends to result in decreased heat resistance (elasticity at elevated temperatures).

[0144]

5 The thermoplastic resin can be used singly or in a combination of a plurality of species.

[0145]

10 The above thermoplastic resin may be mixed with the component (A) and/or (B) by homogeneously dissolving therein, dispersing therein in the form of ground particles, dispersing therein in the form of a solution in a solvent, or the like. From the viewpoint that the resulting curing product readily becomes more transparent, the resin is preferably mixed with the component (A) and/or (B) by homogeneously dissolving
15 therein. In this case, too, the thermoplastic resin may be directly dissolved in the component (A) and/or (B) or homogeneously mixed therewith using a solvent or the like, or the solvent may be removed thereafter to attain a homogeneous dispersion state and/or mixture state.

20 [0146]

When the thermoplastic resin is dispersed, the average particle diameter can be selected at various levels. Preferably, however, the lower limit to the average particle diameter is 10 nm, and the upper limit to the average particle
25 diameter is preferably 10 μ m. The particle system may have a distribution and may be monodisperse or show a plurality of peak particle diameters. In view of the tendency for the curable composition to readily become low in viscosity and show better moldability, the coefficient of variation in particle diameter
30 is preferably not more than 10%.

[0147]

A filler may be added to the curable composition of the invention.

[0148]

35 The above filler may be various ones, for example, silica

type filler such as quartz, fumed silica, precipitated silica, silicic anhydride, fused silica, crystalline silica and ultrafine amorphous silica; inorganic fillers such as silicon nitride, silver powders, alumina, aluminum hydroxide, titanium oxide, glass fibers, carbon fibers, mica, carbon black, graphite, diatomaceous earth, terra alba, clay, talc, calcium carbonate, magnesium carbonate, barium sulfate and inorganic ballon; those fillers in common use in and/or proposed as fillers for the conventional sealing member, such as an epoxy type filler; and the like.

[0149]

As the filler, those low in radioactivity are preferred from the viewpoint that semiconductors or electric materials to be sealed are hardly damaged.

[0150]

The filler may be surface-treated where appropriate. The surface treatment includes alkylation treatment, trimethylsilylation treatment, silicone treatment, treatment with a coupling agent, etc.

[0151]

As examples of the above coupling agent, there may be mentioned a silane coupling agent. The silane coupling agent is not particularly restricted but may be any of those compounds which have, within the molecule, at least one functional group reactive with an organic group and at least one hydrolyzable silyl group. The functional group reactive with an organic group is preferably at least one functional group selected from among epoxy, methacryl, acryl, isocyanate, isocyanurate, vinyl and carbamate groups from the handleability viewpoint. From the viewpoint of curability and adhesive properties, epoxy, methacryl and acryl groups are particularly preferred. The hydrolyzable silyl group is preferably an alkoxysilyl group and, from the reactivity viewpoint, a methoxysilyl and an ethoxysilyl groups are particularly preferred.

[0152]

As preferred examples of the silane coupling agent, there may be mentioned epoxy functional group-containing alkoxy silanes such as 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropyltriethoxysilane, 5 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane and 2-(3,4-epoxycyclohexyl)ethyltriethoxysilane; and methacryl or acryl group-containing alkoxy silanes such as 3-methacryloxypropyltrimethoxysilane, 3-methacryloxypropyltriethoxysilane, 10 3-acryloxypropyltrimethoxysilane, 3-acryloxypropyltriethoxysilane, methacryloxymethyltrimethoxysilane, methacryloxymethyltriethoxysilane, acryloxymethyltrimethoxysilane and 15 acryloxymethyltriethoxysilane.

[0153]

In addition, a process for incorporating a filler may be mentioned. There may also be mentioned, for example, a method of forming a filler in the curable composition which comprises 20 adding a hydrolyzable silane monomer or oligomer, such as an alkoxy silane, acyloxy silane or halogenated silane, or an alkoxide, acyloxy or halide of a metal, such as titanium or aluminum, to the composition of the invention and allowing the filler formation reaction to proceed in the composition or the 25 partial reaction product derived from the composition.

[0154]

Among the above fillers, a silica type filler is preferred from the viewpoint that it makes it difficult for the curing reaction to be inhibited and is highly effective in reducing 30 the coefficient of linear expansion.

[0155]

The above filler preferably has an average particle diameter of not more than 10 μm , more preferably not more than 5 μm , since good permeability to narrow gaps of a sealing member 35 can be easily obtained then.

[0156]

From the better permeability to narrow gaps of a sealing member viewpoint, the proportion of particles having a diameter of 50 μm or larger in the filler is preferably not more than 1% by weight, more preferably not more than 0.1% by weight.

[0157]

The particle diameter distribution of the above filler can be selected in various ways, typically in those ways employed in and/or proposed for the fillers in the conventional sealing members such as the epoxy type ones. For example, particles larger than 24 μm may amount to not less than 15% by weight while particles not larger than 1 μm may amount to not less than 3% by weight.

[0158]

The average particle diameter of the filler and the proportion of particles not smaller than 50 μm in the filler can be determined using a laser microtrack particle size analyzer.

[0159]

The specific surface area of the above filler can be selected at various levels, typically at the levels employed and/or proposed for the fillers in the conventional sealants, such as the epoxy-based ones. For example, levels of not less than 4 m^2/g , not more than 4 m^2/g , not more than 10 m^2/g and so forth may be optionally employed.

[0160]

The specific surface area can be measured using a BET method-based monosorb specific surface area measuring apparatus.

[0161]

The vitrification percentage of the above filler can be selected at various levels, typically at the levels employed and/or proposed for the fillers in the conventional sealing members, such as the epoxy-based ones. For example, levels of not less than 97%, and the like can be optionally employed.

[0162]

The above filler preferably has a spherical shape from the viewpoint that the sealing member viscosity tends to become low.

5 [0163]

The filler may comprise one single species or a combination of two or more species.

[0164]

10 The level of addition of the above filler is not particularly restricted but, from the viewpoint that the linear expansion coefficient reducing effect is high and the composition has good fluidity, the lower limit to the addition level is preferably 30% by weight, more preferably 50% by weight, of the whole composition, and the upper limit to the addition
15 level is preferably 80% by weight, more preferably 70% by weight, of the whole composition.

[0165]

Various methods can be employed for filler incorporation. For better storage stability of the intermediate material of
20 the composition, the method comprising mixing the component (B) with a mixture prepared by mixing the component (A) with the component (C) and the filler is preferred. When the component (A) is mixed with a mixture prepared by mixing the component (B) with the component (C) and/or the filler, the composition
25 may undergo denaturation during storage, etc. since the component (B) has reactivity with the moisture in the environment and/or with the filler in the presence and/or absence of the component (C).

[0166]

30 An antioxidant may be added to the curable composition of the invention. The antioxidant includes those in conventional use, for example citric acid, phosphoric acid, sulfur-containing antioxidants, etc. As the sulfur-containing antioxidants, there may be mentioned
35 mercaptans, mercaptan salts, sulfide carboxylic acid esters,

hindered phenol type sulfides and other sulfides, polysulfides, dithiocarboxylic acid salts, thioureas, thiophosphates, sulfonium compounds, thioaldehydes, thioketones, mercaptals, mercaptols, monothio acids, polythio acids, thioamides, 5 sulfoxides, etc.

[0167]

The above antioxidants may be used singly or two or more of them may be used in combination.

[0168]

10 A radical inhibitor may be added to the composition of the invention. The radical inhibitor includes, for example, phenolic radical inhibitors such as 2,6-di-tert-butyl-3-methylphenol (BHT), 2,2'-methylenebis(4-methyl-6-tert-butylphenol) and 15 tetrakis(methylene-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate)methane, amine type radical inhibitors such as phenyl- β -naphthylamine, α -naphthylamine, N,N'-sec-butyl-p-phenylenediamine, phenothiazine and N,N'-diphenyl-p-phenylenediamine, etc.

20 [0169]

The above radical inhibitors may be used singly or two or more of them may be used in combination.

[0170]

25 An ultraviolet absorber may be added to the composition of the invention. The ultraviolet absorber includes, for example,

2-(2'-hydroxy-3',5'-di-tert-butylphenyl)benzotriazole, bis(2,2,6,6-tetramethyl-4-piperidine) sebacate, and so on.

[0171]

30 The above ultraviolet absorbers may be used singly or two or more of them may be used in combination.

[0172]

To the composition of the invention, other than the above components, there may further be added components typically at 35 the levels employed and/or proposed for the fillers in the

conventional sealants, such as the epoxy-based ones, and colorants, mold release agents, flame retardants, flame retardant auxiliaries, surfactants, antifoaming agents, emulsifiers, leveling agents, cissing preventing agents, ion trapping agents, thixotropic agents, tackifiers, storage stability improving agents, antiozonants, light stabilizers, thickening agents, plasticizers, reactive diluents, antioxidants, heat stabilizers, electric conductivity providing agents, antistatic agents, antiradiation agents, nucleating agents, phosphorus-containing peroxide decomposers, lubricants, pigments, metal inactivators, thermal conductivity providing agents, physical property modifiers and so forth, each within the range within which the object and effect of the invention will not be defeated or weakened.

15 [0173]

The composition of the invention can also be used in the form of a solution in a solvent. The solvent that can be used is not particularly restricted but, for example, there may be specifically mentioned hydrocarbon solvents such as benzene, toluene, hexane and heptane; ether solvents such as tetrahydrofuran, 1,4-dioxane, 1,3-dioxolane and diethyl ether; ketone solvents such as acetone, methyl ethyl ketone and methyl isobutyl ketone; and halogenated solvents such as chloroform, methylene chloride and 1,2-dichloroethane.

25 [0174]

Among these, preferred are toluene, tetrahydrofuran, 1,3-dioxolane, and chloroform.

[0175]

The amount of the solvent to be used can be selected optionally. However, the lower limit to the amount to be used is preferably 0.1 mL, and the upper limit thereto is preferably 10 mL, per gram of the curable composition. When the solvent amount used is small, the effects of the use of a solvent, for example the viscosity reducing effect and the like effect, tends to become difficult to obtain and, when it is large, the solvent

tends to remain in the material, readily causing heat crack and other problems, and tends to become disadvantageous from the cost viewpoint, thus the industrial utility value becomes decrease.

5 [0176]

The above solvents may be used singly or in the form of a mixed solvent composed of two or more species.

[0177]

10 The composition of the invention may be incorporated with an additive for improving various properties of the light-emitting diode. As the above additive, for example, there may be mentioned cerium-activated yttrium aluminum garnet type fluorescent substances which absorb the light from the light emitting element and emit a longer wavelength fluorescent
15 light, and the like fluorescent substances; colorants such as a bluing agent which absorb a specific wavelength; inorganic or organic diffusing agents for diffusing the light such as titanium oxide, aluminum oxide, silica, silicon oxide e.g. silica glass, talc, calcium carbonate, melamine resins, CTU
20 guanamine resins and benzoguanamine resins; thermally conductive fillers such as glass, metal oxide e.g. aluminosilicate, etc. and metal nitrides i.e. aluminum nitride, boron nitride, etc.; etc.

[0178]

25 The additive for improving the light-emitting diode properties may be uniformly contained or may be contained in a gradient manner in the composition. The above filler-containing resin part can be formed as a sealing member of the back side of the light emitting surface by pouring a resin
30 for a sealing member of the front side of the light emitting surface into a mold, and successively pouring a resin containing the above filler. Moreover, the above filler-containing resin part can also be formed by covering a lead terminal by taping both sides after forming a sealing member, and in this condition,
35 immersing the whole lead frame in a tank filled with the above

filler-containing resin in such manner that the lower half of the sealing member of the light-emitting diode being immersed, withdrawing thereof, and drying.

[0179]

5 A light-emitting diode can be produced using the composition of the invention. The light-emitting diode of the invention can be produced by coating the light emitting element with the composition mentioned above.

[0180]

10 The above light emitting element is not particularly restricted, and may be any of the known light emitting elements which can be used in the light-emitting diodes. For example, there may be mentioned those produced by layering a semiconductor material, by any of various methods, for example
15 the MOCVD, HDVPE, and liquid phase growth methods, on a substrate, if necessary provided with a buffer layer such as a GaN, AlN layer, etc.

As the above substrate, various materials can be used, for example sapphire, spinel, SiC, Si, ZnO, GaN single crystals,
20 etc. Among these, the use of sapphire is preferred since GaN with good crystallinity can be formed with ease and the industrial utility is high.

[0181]

The above semiconductor material to be layered includes,
25 for example, GaAs, GaP, GaAlAs, GaAsP, AlGaInP, GaN, InN, AlN, InGaN, InGaAlN, SiC, etc. Among these, nitride compound semiconductors ($\text{In}_x\text{Ga}_y\text{Al}_z\text{N}$) are preferred from the high brightness viewpoint. The above materials may contain an activator, etc.

30 [0182]

As the structure of the above light emitting element, there may be mentioned, for example, the homojunction, heterojunction, and double-hetero structure having a MIS junction, pn junction or PIN junction. The single or multiple
35 quantum well structure may also be employed.

[0183]

The light emitting element may have a passivation layer or have no passivation layer.

[0184]

5 The light emitting element can be provided with electrodes by any of the methods known in the art.

[0185]

10 The electrodes on the light emitting element can be electrically connected with lead terminals and so forth by various methods. As the electrically connecting members, preferably ones showing good ohmic and mechanical connectivity with the electrodes of the light emitting element, etc., for example bonding wires made of gold, silver, copper, platinum, aluminum, or alloys thereof, etc. Conductive adhesives or the
15 like composed of a conductive filler, such as silver or carbon, and of a resin filled therewith may also be used. From the good workability viewpoint, aluminum wires or gold wires are preferably used among them.

[0186]

20 A light emitting element can be thus obtained. In the present invention, any light emitting element showing luminous intensity in the perpendicular direction is not less than 1 cd may be used. When the light emitting element has a luminous intensity in the perpendicular direction of not less than 2 cd,
25 however, the effects of the invention can be produced more significantly and, when the luminous intensity is not less than 3 cd, the effects of the invention are still more significant.

[0187]

30 The emission output of the above light emitting element is not particularly restricted, and any of those may be used. When the light emitting element shows an output of not less than 1 mW at 20 mA, however, the effects of the invention are significant. When the light emitting element shows an output of not less than 4 mW at 20 mA, the effects of the invention
35 are more significant and, when the light emitting element shows

an output of not less than 5 mW at 20 mA, the effects of the invention are still more significant.

[0188]

The above light emitting element that can be used is not particularly restricted, but may be diverse in emission wavelength range, from the ultraviolet to the infrared region. When the element shows a main emission peak wavelength of not longer than 550 nm, however, the effects of the invention are significant.

10 [0189]

Only one of the above light emitting element may be used for monochromic light emission, or a plurality of such elements may be used for monochromic or polychromic light emission.

[0190]

15 The lead terminals to be used in the light-emitting diode of the invention are preferably ones showing good adhesion to electric connection members, such as bonding wires, good electric conductivity, etc. Thus, the lead terminals preferably have an electric resistance of not more than 300 $\mu\Omega\cdot\text{cm}$, more preferably not more than 3 $\mu\Omega\cdot\text{cm}$. As the material of these lead terminals, there may be mentioned, for example, iron, copper, iron-containing copper, tin-containing copper and, further, the products of plating of these with silver, nickel, or the like, etc. The glossiness of these lead
20 terminals may be appropriately adjusted for attaining good spreading of light.
25

[0191]

The light-emitting diode of the invention can be produced by coating the light emitting element with the composition mentioned above. The above-mentioned "coating" includes, within the meaning thereof, not only direct sealing of the light emitting element but also indirect coating. Specifically, the above light emitting element may be directly sealed with the composition of the invention by various methods conventionally
30 in use, or after sealing the light emitting element with known
35

sealing resins such as epoxy resins, silicone resins, acrylic resins, urea resins and imide resins, or glass, and then the above or surround the resultant may be coated with the composition of the invention. Moreover, after sealing the
5 above light emitting element with the composition of the invention, the resultant may be molded with known epoxy resins, silicone resins, acrylic resins, urea resins, imide resins, etc. By these methods, utilizing differences of reflective index and specific gravity, various effects such as lens effect can be
10 provided.

[0192]

Various methods can be applied as the method of sealing. For example, a liquid composition may be poured into a cup, cavity, package hollow, etc., with the light emitting element
15 disposed on the bottom thereof in advance, using a dispenser or by any other method, followed by curing by heating, etc. It is also possible to allowing a solid composition or highly viscous liquid composition to flow by heating, etc., pouring the same into a package hollow or the like in the same manner and causing curing by heating, etc. The above package can be
20 produced using any of various materials, and there may be mentioned, for example, polycarbonate resins, polyphenylene sulfide resins, epoxy resins, acrylic resins, silicone resins, ABS resins, polybutylene terephthalate resins,
25 polyphthalamide resins, etc. Moreover, a method comprising pouring the composition into a molding form in advance, immersing a lead flame or the like fixed with the light emitting element thereto, and then curing the composition can also be applied. Or the sealing layer comprising the composition may
30 be molded and cured by pouring using a dispenser into a form inserted with the light emitting element, transfer molding, or injection molding, etc. It is also possible to simply cure the composition in a liquid or flowage form by dropwise addition in the shape of the light emitting element or coating. The
35 curable composition can also be molded and cured by

mimeographing, screen printing, application via a mask, and the like. Or a method comprising fixing a composition partially cured or cured in a plate-like shape or lens-like shape on the light emitting element can also be applied. Moreover, the composition can be used as a die-bonding agent for fixing the light emitting element to a lead terminal or a package, or as a passivation layer on the light emitting element. Further, it can also be used as a package substrate.

[0193]

10 The shape of the coated portion is not particularly restricted but may be diverse. For example, there may be mentioned lens-like shapes, plate-like shapes, thin film shapes, those shapes described in Japanese Kokai Publication Hei-06-244458, and the like shapes. These shapes may be formed by molding and curing of a composition or by curing of a composition, followed by after-working.

[0194]

20 The light-emitting diode of the invention can be applied in various types. For example, either types such as lump type, SMD type, and tip type can be applied. As a packaging substrate of SMD and tip types, various ones may be used, and there may be mentioned, for example, epoxy resins, BT resins, ceramics, etc.

[0195]

25 Further, various known modes can be applied to the light-emitting diode of the invention. For example, there may be mentioned the mode in which a light-reflecting or light-collecting layer is provided on the back of the light emitting element, the mode in which a complementarily colored portion is provided on the bottom to cope with sealant resin yellowing, the mode in which a thin film capable of absorbing shorter wavelength light than the main emission peak is disposed on the light emitting element, the mode in which the light emitting element is sealed with a soft or liquid sealing member and then the circumference is molded with a hard material, the

30

35

mode in which the light emitting element is sealed with a material containing a fluorescent substance absorbing the light from the light emitting element and emitting a longer wavelength fluorescent light and the circumference is then molded, the mode
5 in which a material containing a fluorescent substance is shaped in advance and then molded together with the light emitting element, the mode in which a sealing member is formed into a special shape to thereby increase the emission efficiency, as described in Japanese Kokai Publication Hei-06-244458, the mode
10 in which the package is provided with a two-stage hollow to reduce the unevenness of brightness, the mode in which the light-emitting diode is immobilized by insertion into a through hole, the mode in which a thin film capable of absorbing a shorter wavelength light than the main emission wavelength is formed
15 on the light emitting element surface, the mode in which the light emitting element is connected with lead members and the like by flip chip bonding or the like using solder bumps, etc., for taking out light in the direction of the substrate and the like modes.

20 [0196]

The light-emitting diode of the invention can be used in various fields of use known in the art. Specifically, such uses include backlights, illumination, light sources for sensors, light sources for gauges in vehicles, signal lamps, display
25 lamps, display devices, light sources for planar light emitters, displays, decorations, various lights, etc.

[0197]

The curable composition of the invention is applicable to various optical materials. The optical materials in the
30 invention refer to materials in general used for transmitting light therein such as visible light, infrared rays, ultraviolet rays, X-rays, and laser.

[0198]

The optical materials in the invention refer to materials
35 in general used for transmitting light therein such as visible

light, infrared rays, ultraviolet rays, X-rays, and laser.

[0199]

There may be mentioned, for example, materials used for liquid crystal display devices such as a color filter overcoat, TFT flattening film, and substrate material; and materials used for light-emitting diodes (LED) such as a sealing member and die-bonding agent. In addition, the sealing member in this specification is a concept including a molding agent or a sealant.

10 [0200]

There may also be mentioned a substrate material, optical waveguide, prism sheet, deflection plate, wave plate, viewing angle correction film, polarizer protection film, a color filter, etc. used in liquid crystal display fields; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0201]

There may also be mentioned a sealing member of LED elements, sealing member of LED, protection film of front face glass, and alternate material for front face glass used in LED display devices; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0202]

There may also be mentioned an antireflective film, optical correction film, housing material, protection film of front face glass, and alternate material for front face glass used in color PDPs (plasma display); and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a substrate material, optical waveguide, prism sheet, deflection plate, wave plate, viewing angle correction film, and polarizer protection film in plasma address liquid crystal (PALC) displays; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a protection film of front face glass, and alternate material for front face glass in

organic electro luminescence displays; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned various film substrates, a protection film of front face glass, and alternate material
5 for front face glass in field emission displays (FED); and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0203]

10 In the optical record fields, there may be mentioned VD (video disk), CD/CD-ROM, CD-R/RW, DVD-R/DVD-RAM, MO/MD, PD (phase change disk), disk substrate material for optical cards, pickup lens, and protection film; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

15 [0204]

In the optical apparatus fields, there may be mentioned a lens material for steel cameras, finder prism, target prism, finder cover, and light-sensitive sensor part; and various coating agents, protection films, sealing members, adhesives,
20 etc. used for them. There may also be mentioned a photographing lens for video cameras, and finder; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a projection lens and protection film for projection TVs; and various coating agents,
25 protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a lens material and various films for optical sensing apparatus; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

30 [0205]

In the optical part fields, there may be mentioned a fiber material for the peripheral of optical switch, lens, waveguide, element in optical communication systems; and various coating agents, protection films, sealing members, adhesives, etc. used
35 for them. There may also be mentioned an optical fiber material

and ferrule for the peripheral of optical connectors; and various coating agents, protection films, sealing members, adhesives, etc. used for them. As optical passive parts and optical circuit parts, there may be mentioned a lens and waveguide; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a substrate material and fiber material for the periphery of opto-electronic integrated circuit (OEIC); and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0206]

In the optical fiber fields, there may be mentioned sensors for industrial application such as lightning for ornament displays and light guide, optical fibers for connecting communication infrastructure and domestic digital equipment such as a display and indicator; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0207]

In the semiconductor integrated circuit peripheral materials, there may be mentioned resist a material of micro lithography for LSI and super LSI materials.

[0208]

In the car and transport plane fields, there may be mentioned lump materials such as for headlight, taillight and indoor lamp for cars, a lamp reflector, a lamp lens, various interior and exterior products such as an exterior and interior panel, and glass substitute; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned an exterior part and glass substitute for railway vehicles; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned an exterior part and glass substitute for airplanes; and various coating agents, protection films, sealing members, adhesives, etc. used for

them.

[0209]

In the architectural fields, there may be mentioned a glass intermediate film, glass substitute, and solar cell peripheral material; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0210]

For agriculture, there may be mentioned a coating film for green houses.

10 [0211]

As the optical and electric functional organic materials for the next generation, there may be mentioned an organic EL element peripheral material, organic photorefractive element, optical amplification element which is a light-light conversion device, optical calculation element, substrate material for the peripheral of organic solar cells, fiber material, sealing member of element; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0212]

20 As a process for curing the curable composition of the invention, the reaction may be allowed to proceed by mere mixing or by heating. From the viewpoint that the reaction rate is rapid and materials generally high in heat resistance are readily obtainable, the method comprising heating to allow the reaction to proceed is preferred.

25 [0213]

The reaction temperature may be selected at various levels. However, the usable temperature is 30 to 300°C, more preferably 100 to 250°C, still more preferably 150 to 200°C. At lower reaction temperatures, the reaction time for the reaction to proceed to a satisfactory extent becomes long, whereas at higher temperatures, molding process tends to be difficult.

[0214]

35 The reaction may be carried out at a constant temperature,

or the temperature may be varied stepwise or continuously according to need. The reaction is preferably carried out with the temperature varied stepwise or continuously, not at a constant temperature, because the cured product to be obtained
5 tends to be strain-free and uniform.

[0215]

The reaction time can also be selected at various levels. The reaction is preferably carried out at lower temperature for comparatively long time, not at elevated temperature for
10 comparatively short time, because the cured product to be obtained tends to be strain-free and uniform.

[0216]

The pressure during reaction also can be selected at various levels according to need. Thus, the reaction can be
15 carried out at ordinary pressure, at an elevated pressure or under reduced pressure. The reaction is preferably carried out under reduced pressure because vaporous components to be produced are easily eliminated by hydrolysis condensation.

[0217]

20 The shapes of the optical material obtained by curing is not particularly restricted since various shapes are applicable according to its application, and include, for example, film, sheet, tube, rod, coat film, and bulk shapes.

[0218]

25 Molding methods also include various methods including conventional methods for molding thermoplastic resins. For example, molding methods such as casting method, press method, cast molding method, transfer molding method, coating method, and RIM method can be applied. As molding tools, polished glass,
30 hard stainless polished board, polycarbonate board, polyethylene terephthalate board, polymethyl methacrylate board, etc. can be applied. Moreover, for improving mold-releasing ability with the molding tool, it is possible to use a polyethylene terephthalate film, polycarbonate film,
35 polyvinyl chloride film, polyethylene film,

polytetrafluoroethylene film, polypropylene film, polyimide film, etc.

[0219]

In the molding, various treatments may be applied according to need. For example, such treatments can also be applied as one comprising defoaming the composition or partly-reacted composition by centrifugation, reduced pressure, etc. for inhibiting generation of a void in the step of the molding, or one comprising releasing pressure once in the step of pressing.

[0220]

[Examples]

The following examples and comparative examples illustrate the present invention. They are, however, by no means limitative of the scope of the present invention. (Synthesis Example 1)

A 5-L separable flask was charged with 1.8 kg of toluene and 1.44 kg of 1,3,5,7-tetramethylcyclotetrasiloxane, and heated until the inside temperature arrived at 104°C. Thereto was added dropwise a mixture of 200 g of triallyl isocyanurate, 1.44 mL of platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) and 200 g of toluene. The resulting mixture was heated under reflux in an oil bath at 120°C for 7 hours. Then, 1.7 g of 1-ethynyl-1-cyclohexanol was added. The unreacted portion of 1,3,5,7-tetramethylcyclotetrasiloxane and the toluene were distilled off under reduced pressure. ¹H-NMR revealed that the product was the reaction product resulting from part of the SiH groups of 1,3,5,7-tetramethylcyclotetrasiloxane with triallyl isocyanurate (hereinafter referred to as partial reaction product A, SiH value: 8.2 mmol/g, allyl value: 0.12 mmol/g).

(Example 1)

A mixture (mixture A) was prepared by mixing up 10.3 g of triallyl isocyanurate, 2.75 g of diallyl monoglycidyl

isocyanurate (20% by weight in component (A)), 93 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum), 310 mg of ALCH-TR (Kawaken Fine Chemicals Co., Ltd.). B mixture (mixture B) was prepared by mixing up 17.96 g of the partial reaction product (A) prepared in Synthesis Example 1, 93 mg of 1-ethynyl-1-cyclohexanol, and 1.55 g of γ -glycidoxypropyltrimethoxysilane with stirring. After mixing up the above mixtures A and B with stirring and degassing, the mixture was poured into a cell produced by inserting a 3-mm-thick silicone rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour and 150°C for 1 hour, in that order, to obtain a transparent and hard molding.

15

(Example 2)

A mixture (mixture A) was prepared by mixing up 7.23 g of triallyl isocyanurate, 7.70 g of diallyl monoglycidyl isocyanurate (50% by weight in component (A)), 99 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum), 329 mg of ALCH-TR (Kawaken Fine Chemicals Co., Ltd.). B mixture (mixture B) was prepared by mixing up 17.96 g of the partial reaction product (A) prepared in Synthesis Example 1, 99 mg of 1-ethynyl-1-cyclohexanol, and 1.64 g of γ -glycidoxypropyltrimethoxysilane with stirring. After mixing up the above mixtures A and B, the mixture was cured under the same condition as in Example 1 to obtain a transparent and hard molding.

30

(Example 3)

A mixture (mixture A) was prepared by mixing up 7.23 g of triallyl isocyanurate, 7.70 g of diallyl monoglycidyl isocyanurate (51% by weight in component (A)), 99 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum), 164 mg of trimethyl bprate. B mixture

35

(mixture B) was prepared by mixing up 17.96 g of the partial reaction product (A) prepared in Synthesis Example 1, 99 mg of 1-ethynyl-1-cyclohexanol, and 822 mg of γ -glycidoxypopyltrimethoxysilane with stirring. After mixing up the above mixtures A and B, the mixture was cured under the same condition as in Example 1 to obtain a transparent and hard molding.

(Comparative Example 1)

10 A mixture (mixture A) was prepared by mixing up triallyl isocyanurate (12.04 g), 90 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum), and 150 mg of ALCH-TR (Kawaken Fine Chemicals Co., Ltd.). B mixture (mixture B) was prepared by mixing up 17.96 g of the
15 partial reaction product (A) prepared in Synthesis Example 1, 90 mg of 1-ethynyl-1-cyclohexanol, 750 mg of γ -glycidoxypopyltrimethoxysilane. After mixing up the above mixtures A and B with stirring and degassing, the mixture was poured into a cell produced by inserting a 3-mm-thick silicone
20 rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour and 150°C for 1 hour, in that order, to obtain a transparent and hard molding.

25 (Measurement Example 1)

 For the samples obtained in Examples 1 to 3 and Comparative Example 1, tensile elasticity and tensile elongation were evaluated. The test samples were prepared as follows. The plate-shaped curing products of 3 mm in thickness were cut into
30 rectangles of 6 x 55 x 3 mm, and near the center of longitudinal sides thereof were notched from both sides in U shape of $r=1.0$ mm and depth of 1 mm. The measurement was carried out using an autograph placed under the condition of 50% RH at 23°C, and the distance between the chucks was set at 15 mm. The U-notch
35 parts of the test samples were placed near the center of the

chucks, both ends thereof were pinched with the chucks, and tensile test was carried out at a tensile speed of 1 mm/min. The obtained results are shown in the following Table.

[0221]

5 [Table 1]

	Tensile characteristics	
	strength, MPa	elongation, %
Example 1	28	4
Example 2	30	5
Example 3	31	6
Compar.Ex. 1	26	6

10 From table 1, it is found that the curing product of the invention has high tensile strength and elongation, as well as high toughness.

15

(Measurement Example 2)

For the samples obtained in Examples 1 to 3 and Comparative Example 1, the light resistance was evaluated at the initial stage and after 70 hours irradiation by a xenon weatherometer. The light transmittance at a wavelength of 470 nm was evaluated before and after the testing. The test was carried out using Suga Test Instruments' super xenon weatherometer (18 minutes of raining + 1 hour and 42 minutes of irradiation) as evaluation equipment under the following test conditions for 70 hours: irradiation illuminance 180 W/m², black panel temperature 63°C, and humidity 50%. As transmittance measuring equipment, spectrophotometer U-3300 manufactured by Hitachi, Ltd. was used. The light transmittance of the curing product obtained in the Examples and Comparative Examples are shown below.

30 [0222]

[Table 2]

35

	Light transmittance at 470 nm. %	
	Initial stage	After light resistance test
Example 1	88	89
Example 2	85	86
Example 3	89	88
Compar.Ex.1	89	88

From table 2, it is found that the curing products of the invention have high light resistance as high as those of the curing products obtained in Comparative Examples.

(Example 4)

The sheet-shaped curing product produced in Example 1 is cut into an appropriate form, and fixed on a light-transmitting window disposed on a metal cap for can type. Meanwhile, a light emitting element having a double-hetero structure in which an Si- and Zn-doped InGaN active layer formed on a sapphire substrate by MOCVD (Metal Organic Chemical Vapor Deposition) method being sandwiched with n- and p-type AlGaN clad layers is prepared. Successively, this light emitting element is mounted on a metal stem for can type, and p- and n-electrodes were wire-bonded with an Au line to the respective leads. This is air-sealed with the above metal cap for can type. In this manner, a can type light-emitting diode can be produced.

(Example 5)

On a washed sapphire substrate, by MOCVD (Metal Organic Chemical Vapor Deposition) method, the below-mentioned layers are laminated in the following order: n-type GaN layer which is an undoped nitride semiconductor; GaN layer which is to be an n-type contact layer formed with an Si-doped n-type electrode; n-type GaN layer which is an undoped nitride semiconductor; GaN layer which is to be a barrier layer constituting a light emitting layer; InGaN layer constituting a well layer; GaN layer which is to be a barrier layer (quantum

well structure); AlGaIn layer as a p-type clad layer doped with Mg on a light emitting layer; and GaIn layer which is an Mg-doped p-type contact layer. By etching, the surfaces of p- and n-contact layers are exposed on the same side of the nitride semiconductor on the sapphire substrate. On the respective contact layers, Al is deposited by a sputtering method to form positive and negative electrodes. A scribing line was drawn on the prepared semiconductor wafer, and then the wafer is divided by external pressure to form a light emitting element.

10 [0223]

On the bottom surface of a silver-plated mount lead cup constituted of iron-containing copper, the above light emitting element is die-bonded using an epoxy resin composition as a die bond resin. The resultant is heated at 170°C for 75 minutes to cure the epoxy resin composition to fix the light emitting element. Next, the positive and negative electrodes of the light emitting element, the mount lead and inner lead are wire-bonded with an Au line to gain electric access.

[0224]

20 A curable composition prepared in the same manner as Example 1 is poured into a casting case, which is a bombshell-shaped molding form. Parts of the mount lead and inner lead, the cup of which is disposed with the above light emitting element, are inserted into the casting case to carry out an initial curing at 100°C for 1 hour. The light-emitting diode is withdrawn from the casting case and cured at 120°C for 1 hour under nitrogen atmosphere. Thereby, a lump type light-emitting diode such as a bombshell-shaped one can be produced.

30

(Example 6)

A curable composition and a light-emitting diode are produced by the method described in Example 1.

[0225]

35 A pair of copper foil patterns is formed on a glass epoxy

resin by etching to produce a substrate having lead electrodes. A light emitting element is die-bonded on the glass epoxy resin using an epoxy resin. The respective electrodes of the light emitting element and respective lead electrodes are wire-bonded
5 with an Au line to gain electric access. On the substrate, a glass epoxy resin having a through hole is fixed and disposed using an epoxy resin to function as a mask and sidewall. In this condition, the resultant is disposed in vacuum equipment, the curable composition is dispensed on the glass epoxy resin
10 substrate disposed with the light emitting element, and the curable composition is filled into a cavity utilizing the through hole. In this condition, the composition is cured at 100°C for 1 hour, and further at 150°C for 1 hour. By dividing with every light-emitting diode tip, a tip type light-emitting
15 diode can be produced.

(Example 7)

A curable composition and a light-emitting diode are produced by the method described in Example 1.

20 [0226]

By insert molding, a package of a tip type light-emitting diode is formed using a PPS resin. Inside of the package, an aperture to be disposed with a light emitting element is equipped and a silver-plated copper plate is placed as an
25 external electrode. A light emitting element is fixed by die bonding inside the package using an epoxy resin. An Au line, which is a conductive wire, is electrically connected to the respective electrodes of the light emitting element and the respective external electrodes disposed in the package by wire
30 bonding. Inside of the package aperture, a curable composition is filled as a sealing member. In this condition, the composition is cured at 100°C for 1 hour, and further at 150°C for 1 hour. In this manner, a tip type light-emitting diode can be produced.

35 [0227]

[Effect of the Invention]

The curable composition of the invention used for a light-emitting diode is high in toughness and good in light resistance, it can be used as a sealing member for
5 light-emitting diodes.

10

15

20

25

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35

[Document Name] Abstract

[Abstract]

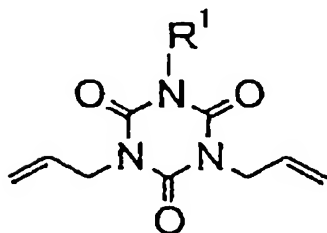
[Subject] The present invention provides a curable composition providing a curing product having high toughness and good light resistance, which may be used in a sealing agent for a light-emitting diode.

[Means for Solving]

A curable composition which contains

(A) a compound represented by the general formula (I):

[Chemical 1]



in the formula, R¹ represents an organic group which does not contain a functional group subjectable to hydrosilylation reaction,

(B) a silicon compound having at least two SiH groups in each molecule, and

(C) a hydrosilylation catalyst.

A curing product having high toughness can be obtained by comprising (A) the compound represented by the formula (I).

[Selective Figure] none

VERIFICATION OF TRANSLATION



I, Fumio Akiyama of MT-2 BLDG., 5-36, Miyahara 3-chome, Yodogawa-ku, Osaka-shi, Osaka 532-0003 JAPAN, hereby declare that I am conversant with the Japanese and English languages and that I am the translator of the documents attached and certify that to the best of my knowledge and belief the following is a true and correct English translation of the Japanese Patent Application No. 2002-125947 in the name of KANEKA CORPORATION.

Dated this 16th day of May, 2007

A handwritten signature in cursive script, appearing to read "Fumio Akiyama", written over a horizontal line.

Fumio Akiyama

Japanese Patent Application No.2002-125947 Filed on April 26, 2002

1

[Document Name] Patent Application
[Filing Number] OSK-4749
[To] Commissioner, Patent Office
[Inventor]
[Address or Residence] A101, Kozenryo, 2-23, Torikainishi
5-chome, Settsu-shi, Osaka
[Name] TUMURA Manabu
[Inventor]
[Address or Residence] D202, Kozenryo, 2-23, Torikainishi
5-chome, Settsu-shi, Osaka
[Name] IDE, Masahito
[Inventor]
[Address or Residence] 5-35-505, Torikainishi 5-chome,
Settsu-shi, Osaka
[Name] OUCHI, Katsuya
[Applicant]
[Identification Number] 000000941
[Name] KANEKA CORPORATION
[Representative] TAKEDA Masatoshi
[Indication of Fee]
[Number of Deposit Ledger] 005027
[The Amount of Payment] 21,000 yen
[List of Attached Documents]
[Document Name] Description 1
[Document Name] Abstract 1
[Necessity of Proof] Needed

[Document Name] Specification

[Title of the Invention] HARDENABLE COMPOSITION, HARDENING PRODUCT, PROCESS FOR PRODUCING THE SAME AND LIGHT EMITTING DIODE SEALED WITH THE HARDENING PRODUCT

5 [Scope of Claims for Patent]

[Claim 1] A curable composition which contains, as essential components,

(A) an organic backbone-based compound containing at least two carbon-carbon double bonds reactive with a SiH group
10 in each molecule,

(B) a silicon compound having at least two SiH groups in each molecule,

(C) a hydrosilylation catalyst,

(D) a silane coupling agent and/or an epoxy
15 group-containing compound, and

(E) a borate ester.

[Claim 2] The curable composition according to Claim 1,

wherein the component (D) is a silane coupling agent having at least one functional group selected from the group
20 consisting of epoxy, methacryl, acryl, isocyanate, isocyanurate, vinyl and carbamate group and hydrolyzable silyl group in each molecule.

[Claim 3] The curable composition according to Claim 1,

wherein the component (D) is a silane coupling agent
25 having an epoxy group and a hydrolyzable silyl group in each molecule.

[Claim 4] The curable composition according to any one of Claims 1 to 3,

wherein the component (E) is at least one species selected
30 from the group consisting of trinormaloctadecyl borate, trinormaloctyl borate, trinormalbutyl borate, triisopropyl borate, trinormalpropyl borate, triethyl borate and trimethyl borate.

[Claim 5] The curable composition according to any one of Claims
35 1 to 4,

wherein the component (A) is triallyl isocyanurate and the component (B) is a reaction product from 1,3,5,7-tetramethylcyclotetrasiloxane and triallyl isocyanurate.

5 [Claim 6] A curing product

which is obtainable by curing the curable composition according to any one of Claims 1 to 5.

[Claim 7] A light-emitting diode

10 which is sealed with the curing product according to Claim 6.

[Detailed Description of the Invention]

[0001]

[Technical Field of the Invention]

15 The present invention relates to a curable composition, more specifically a curable composition having high toughness and low degrees of coloration at high temperatures, a process for producing the same, a curing product thereof, and a light-emitting diode sealed with the curing product.

[0002]

20 [Prior Art]

In the prior art, it is known that a curable composition comprising an organosilicon compound, which is an addition reaction-curable (hydrosilylated) silicone having epoxy group- and alkoxy group-bound silicon atoms, and an
25 organoaluminum compound shows good adhesive properties (Japanese Kokai Publication Hei-08-183934). However, there is problems, i.e. the compound has an insufficient strength and elongation though having good adhesiveness, and low degrees of coloration at high temperatures.

30 As the optical material including a sealing material for light-emitting diodes (LED), with the change of using environment in recent years, those having not only high optical transparency but also high toughness and low degrees of coloration at high temperatures have been also demanded.

35 [0003]

[Subject which the Invention is to Solve]

Accordingly, the object of the present invention is to provide a curable composition, more specifically a curable composition having high toughness and low degrees of coloration at high temperatures, a process for producing the same, a curing product thereof, and a light-emitting diode sealed with the curing product.

[0004]

[Means for Solving the Problems]

10 The present inventors made intensive investigations to accomplish the above object and, as a result, found that it is possible to accomplish the above object by using a curable composition which contains, as essential components, (A) an organic backbone-based compound containing at least two
15 carbon-carbon double bonds reactive with a SiH group in each molecule, (B) a silicon compound having at least two SiH groups in each molecule, (C) a hydrosilylation catalyst, (D) a silane coupling agent and/or an epoxy group-containing compound, and (E) a borate ester. Based on this finding, they have now
20 completed the present invention.

[0005]

Thus, the present invention is:

A curable composition which contains, as essential components,

25 (A) an organic backbone-based compound containing at least two carbon-carbon double bonds reactive with a SiH group in each molecule,

 (B) a silicon compound having at least two SiH groups in each molecule,

30 (C) a hydrosilylation catalyst,

 (D) a silane coupling agent and/or an epoxy group-containing compound, and

 (E) a borate ester (Claim 1),

 The curable composition according to Claim 1,
35 wherein the component (D) is a silane coupling agent

having at least one functional group selected from the group consisting of epoxy, methacryl, acryl, isocyanate, isocyanurate, vinyl and carbamate group and hydrolyzable silyl group in each molecule (Claim 2).

5 The curable composition according to Claim 1,
 wherein the component (D) is a silane coupling agent having an epoxy group and a hydrolyzable silyl group in each molecule (Claim 3).

10 The curable composition according to any one of Claims 1 to 3,

 wherein the component (E) is at least one species selected from the group consisting of trinormaloctadecyl borate, trinormaloctyl borate, trinormalbutyl borate, triisopropyl borate, trinormalpropyl borate, triethyl borate and trimethyl
15 borate (Claim 4).

 The curable composition according to any one of Claims 1 to 4,

 wherein the component (A) is triallyl isocyanurate and the component (B) is a reaction product from
20 1,3,5,7-tetramethylcyclotetrasiloxane and triallyl isocyanurate (Claim 5).

 A curing product

 which is obtainable by curing the curable composition according to any one of Claims 1 to 5 (Claim 6).

25 A light-emitting diode

 which is sealed with the curing product according to Claim 6 (Claim 7).

[0006]

[Embodiment of carrying out the Invention]

30 In the following, the present invention is described in detail.

 First, the component (A) of the present invention is described.

[0007]

35 The component (A) is not particularly restricted but may

be any organic compound having at least two carbon-carbon double bonds reactive with a SiH group in each molecule. The above organic compound is preferably a compound other than a polysiloxane-organic block copolymer, a polysiloxane-organic graft copolymer or a like siloxane unit (Si-O-Si)-containing one, and a compound comprising C, H, N, O, S and/or halogen, as exclusive constituent elements. Siloxane unit-containing compounds have gas permeability and cissing problems.

[0008]

10 The sites of occurrence of the carbon-carbon double bonds reactive with a SiH group are not particularly restricted but may be anywhere within the molecule.

[0009]

15 The component (A) compound may be classified as an organic polymer type compound or as an organic monomer type compound.

[0010]

20 As the organic polymer type compounds, there may be mentioned, for example, polyether type, polyester type, polyarylate type, polycarbonate type, saturated hydrocarbon type, unsaturated hydrocarbon type, polyacrylic ester type, polyamide type, phenol-formaldehyde type (phenol resin type), and polyimide type compounds, etc.

[0011]

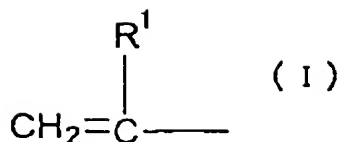
25 As the organic monomer type, there may be mentioned, for example, phenol type, bisphenol type, benzene, naphthalene or like aromatic hydrocarbon type compounds; aliphatic (e.g. straight chain, alicyclic) hydrocarbon type compounds; heterocyclic type compounds; mixtures of these, etc.

[0012]

30 The carbon-carbon double bonds reactive with a SiH group in the component (A) are not particularly restricted but, from the reactivity viewpoint, those groups represented by the following general formula (I):

[0013]

35 [Chemical 1]

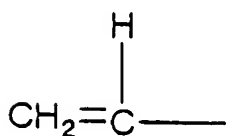


5 wherein R^1 represents a hydrogen atom or a methyl group, are preferred. From the material availability viewpoint, the group represented by

[0014]

[Chemical 2]

10



is particularly preferred.

15

[0015]

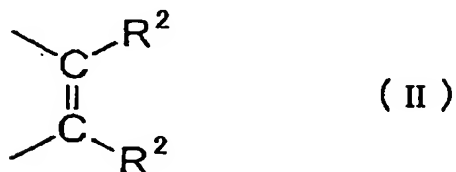
Moreover, preferred as the carbon-carbon double bonds reactive with a SiH group in the component (A) are alicyclic groups having a partial structure represented by the following general formula (II):

20

[0016]

[Chemical 3]

25



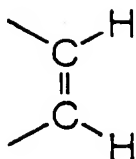
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wherein each R^2 represents a hydrogen atom or a methyl group, in the ring structure, since high heat resistance can be given to curing products. Among them, from the material availability viewpoint, alicyclic groups having a partial structure represented by the following formula

[0017]

[Chemical 4]

35

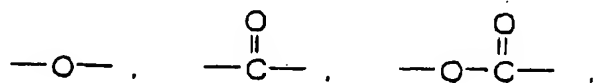


5 in the ring structure are preferred.

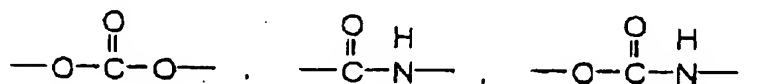
The carbon-carbon double bonds reactive with a SiH group each may be directly bonded to the skeletal portion of the component (A) or covalently bonded thereto via a bivalent or polyvalent substituent. The above bivalent or polyvalent
 10 substituent is not particularly restricted but preferably a substituent containing 0 to 10 carbon atoms, and more preferably one containing C, H, N, O, S and halogen, as exclusive constituent elements. Examples of the above-mentioned substituent are:

15 [0018]

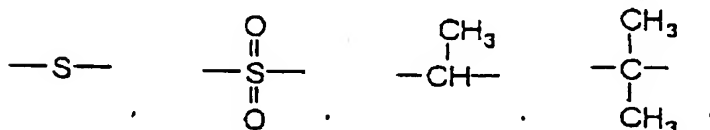
[Chemical 5]



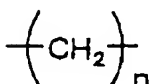
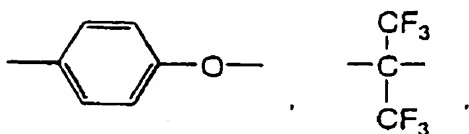
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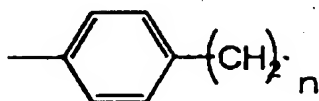


30



35

(n representing a number of 1 to 10),

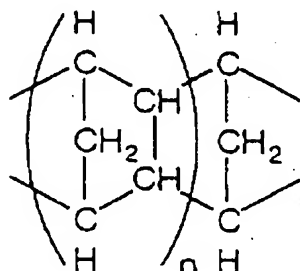


5 (n representing a number of 0 to 4), and

[0019]

[Chemical 6]

10



15 (n representing a number of 0 to 4), etc. Two or more of the bivalent or polyvalent substituents recited above may be covalently bonded together to form one bivalent or polyvalent substituent.

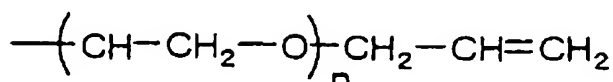
[0020]

20 As examples of the group covalently bonded to the skeletal portion, there may be mentioned vinyl, allyl, methallyl, acryl, methacryl, 2-hydroxy-3-(allyloxy)propyl, 2-allylphenyl, 3-allylphenyl, 4-allylphenyl, 2-(allyloxy)phenyl, 3-(allyloxy)phenyl, 4-(allyloxy)phenyl, 2-(allyloxy)ethyl, 25 2,2-bis(allyloxymethyl)butyl, 3-allyloxy-2,2-bis(allyloxymethyl)propyl,

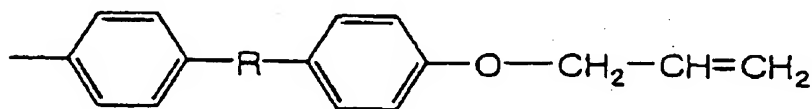
[0021]

[Chemical 7]

30

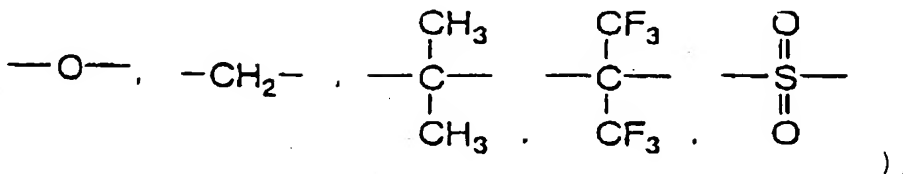


(n representing a number satisfying the relation $5 \geq n \geq 2$),

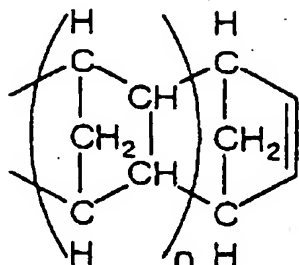


35

(R representing a bivalent group selected from among



5



10

(n representing a number of 0 to 4), and the like.

[0022]

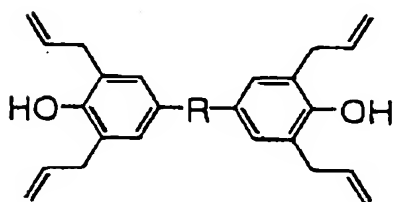
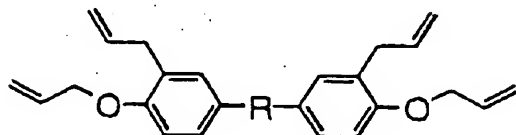
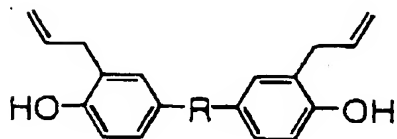
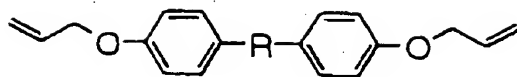
15 As specific examples of the component (A), there may be mentioned diallyl phthalate, triallyl trimellitate, diethylene glycol bis(allyl carbonate), trimethylolpropane diallyl ether, pentaerythritol triallyl ether, 1,1,2,2-tetraallyloxyethane, diallylidenepentaerythritol, 20 triallyl cyanurate, triallyl isocyanurate, 2,2-bis(4-hydroxy cyclohexyl)propane diallyl ether, 1,2,4-trivinylcyclohexane, divinylbenzenes (purity 50 to 100%, preferably 80 to 100%), divinylbiphenyl, 1,3-diisopropenylbenzene, 1,4-diisopropenylbenzene, and oligomers thereof, 25 1,2-polybutadiene (1,2 content 10 to 100%, preferably 50 to 100%), novolak phenol allyl ether, allylated poly(phenylene oxide),

[0023]

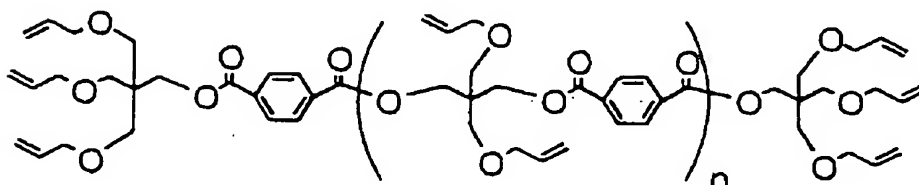
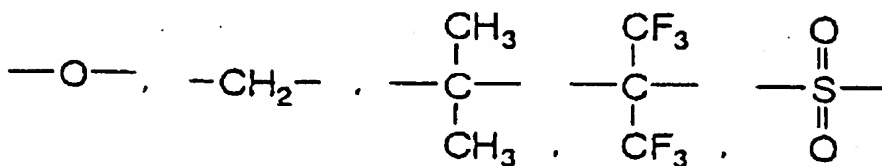
[Chemical 8]

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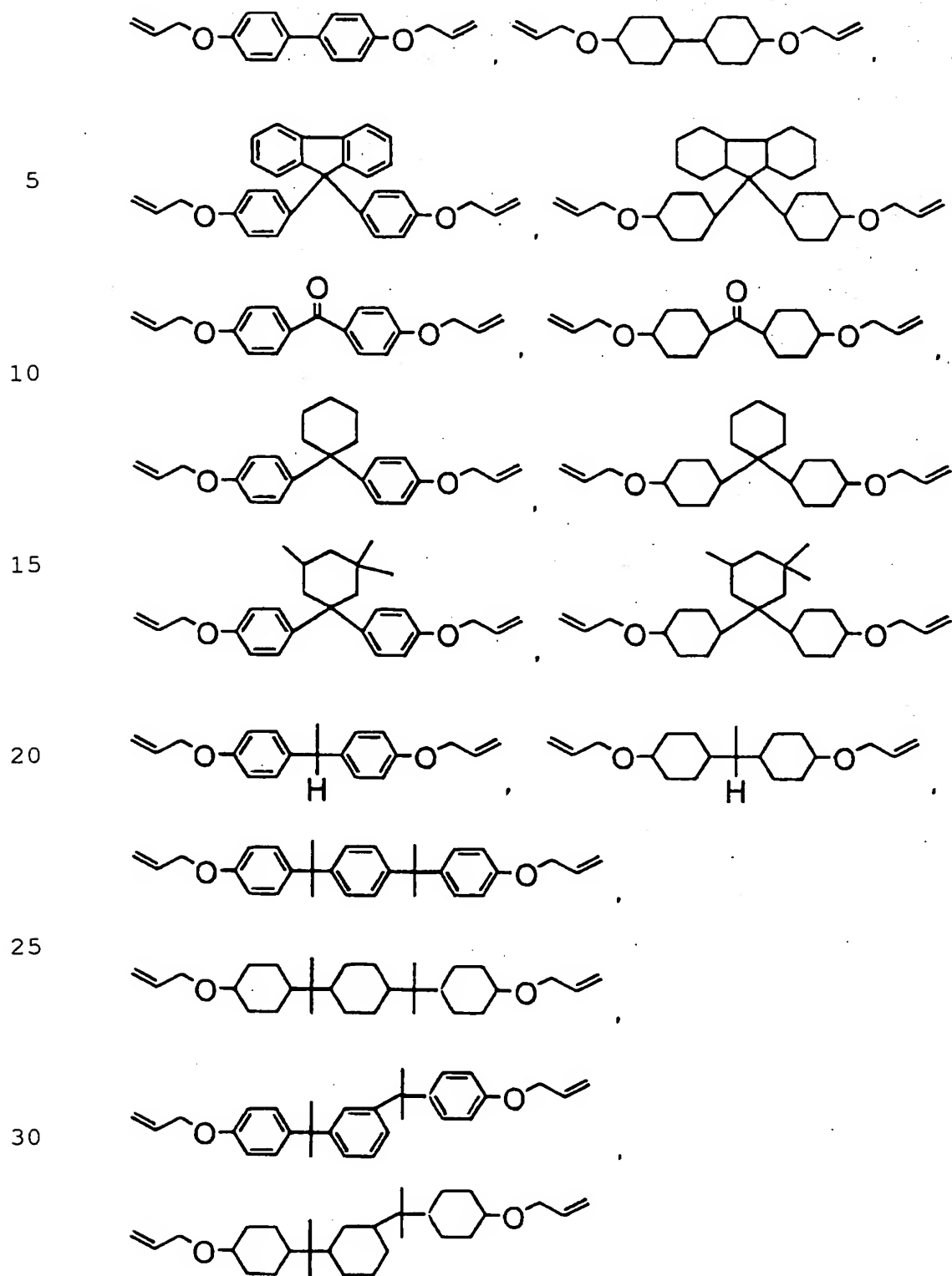


(R representing a bivalent group selected from among


$$(n \div 1)$$

[0024]

[Chemical 9]



and, further, products derived from known epoxy resins by allyl

group substitution for part or all of the glycidyl groups of the resins.

[0025]

Also usable as the component (A) are low-molecular-weight compounds which can hardly be expressed dividedly in terms of a skeletal portion and an alkenyl group as mentioned above. As specific examples of the above low-molecular-weight compounds, there may be mentioned aliphatic linear polyene compounds such as butadiene, isoprene, octadiene and decadiene, alicyclic polyene compounds such as cyclopentadiene, cyclohexadiene, cyclooctadiene, dicyclopentadiene, tricyclopentadiene and norbornadiene, substituted alicyclic olefin compounds such as vinylcyclopentene and vinylcyclohexene, etc.

[0026]

From the viewpoint of the possibility of achieving further improvements in heat resistance, it is preferred that the component (A) contain the carbon-carbon double bonds reactive with a SiH group in an amount of not less than 0.001 mole, more preferably not less than 0.005 mole, still more preferably not less than 0.008 mole, per gram of the component (A).

[0027]

As for the number of carbon-carbon double bonds reactive with a SiH group in the component (A), it is only required that the number be at least 2 per molecule on average. For achieving further improvements in mechanical strength, the number is preferably more than 2, more preferably not less than 3. If the number of carbon-carbon double bonds reactive with a SiH group in the component (A) is 1 or less per molecule, the component (A), upon reacting with the component (B), will only give a graft structure but will fail to give a crosslinked structure.

[0028]

From the good reactivity viewpoint, the component (A) preferably contains at least one vinyl group, more preferably

two or more vinyl groups, in each molecule. From the viewpoint of tendency toward better storage stability, it preferably contains not more than 6 vinyl groups, more preferably not more than 4 vinyl groups, in each molecule.

5 [0029]

From the viewpoint of high mechanical heat resistance and from the viewpoint of less stringiness, good moldability, and handleability, the component (A) preferably has a molecular weight lower than 900, more preferably lower than 700, still
10 more preferably lower than 500.

[0030]

For attaining uniform admixture with other components and good workability, the component (A) preferably has a viscosity at 23°C of lower than 1000 P, more preferably lower than 300
15 P, still more preferably lower than 30 P. The viscosity can be determined using an E type viscometer.

[0031]

From the viewpoint of inhibition of coloration (in particular yellowing), the component (A) is preferably low in
20 content of a compound having a phenolic hydroxyl group and/or a group derived from a phenolic hydroxyl group and, more preferably, it is free of any compound having a phenolic hydroxyl group and/or a group derived from a phenolic hydroxyl group. The term "phenolic hydroxyl group" so referred to herein
25 means a hydroxyl group directly bound to an aromatic hydrocarbon moiety, for example a benzene, naphthalene or anthracene ring, and the "group derived from a phenolic hydroxyl group" means a group resulting from substitution of an alkyl (e.g. methyl, ethyl, etc.), alkenyl (e.g. vinyl, allyl, etc.), acyl (e.g.
30 acetoxyl, etc.) group for the hydrogen atom of the above phenolic hydroxyl group.

[0032]

From the viewpoint of preferable optical characteristics such as low complex reflective index and low optical elastic
35 coefficient as well as preferable weather resistance, the

component (A) preferably contains the aromatic ring component weight ratio of not more than 50% by weight, more preferably 40% by weight, and still more preferably 30% by weight. Most preferably, the component (A) does not contain any aromatic hydrocarbon ring.

[0033]

Preferred as the component (A) from the viewpoint of less coloration, high optical transparency and high light resistance of the curing product obtained are vinylcyclohexene, dicyclopentadiene, triallyl isocyanurate, 2,2-bis(4-hydroxycyclohexyl)propane diallyl ether, and 1,2,4-trivinylcyclohexane. Particularly preferred are triallyl isocyanurate, 2,2-bis(4-hydroxycyclohexyl)propane diallyl ether, and 1,2,4-trivinylcyclohexane.

[0034]

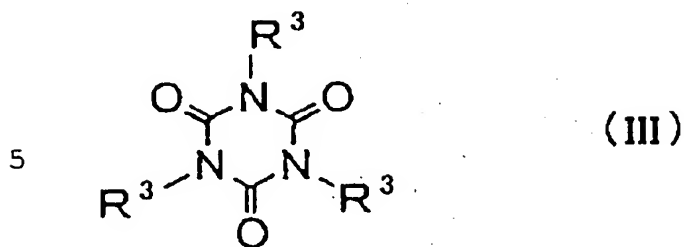
The component (A) may have other reactive group. As the above reactive group, there may be mentioned the following groups, for example: epoxy, amino, radical-polymerizable unsaturated group, carboxyl, isocyanate, hydroxyl, alkoxysilyl group and so on. In cases where the component (A) has the above functional group, the curable composition obtained tends to show increased adhesive properties and the strength of the resulting curing product tends to become high. An epoxy group is preferred among those functional groups in view of the tendency for the resulting curable composition to have better adhesive properties. In view of the tendency for the resulting curing product to become higher in heat resistance, it is preferred that the component (A) has not less than one reactive group mentioned above per molecule on average.

[0035]

From the high heat resistance and transparency viewpoint, the component (A) is particularly preferably a triallylisocyanurate and derivatives thereof represented by the following general formula (III):

[0036]

[Chemical 10]



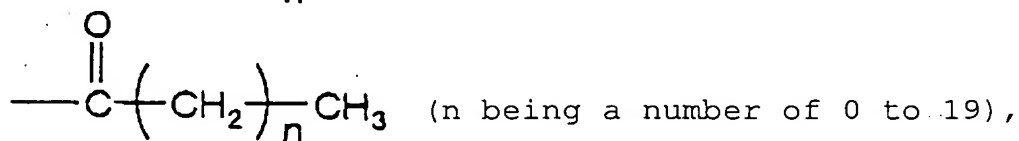
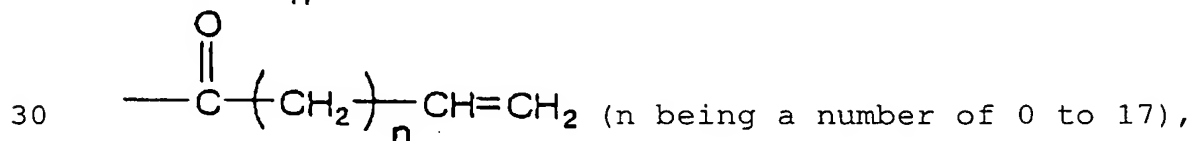
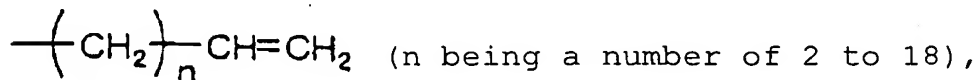
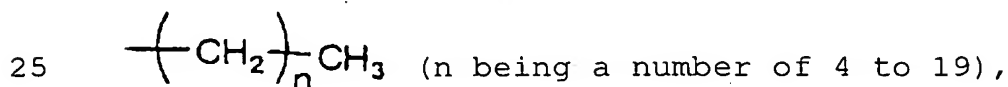
10 wherein each R^3 represents a univalent organic group containing 1 to 50 carbon atoms and the three R^3 groups may be the same or different.

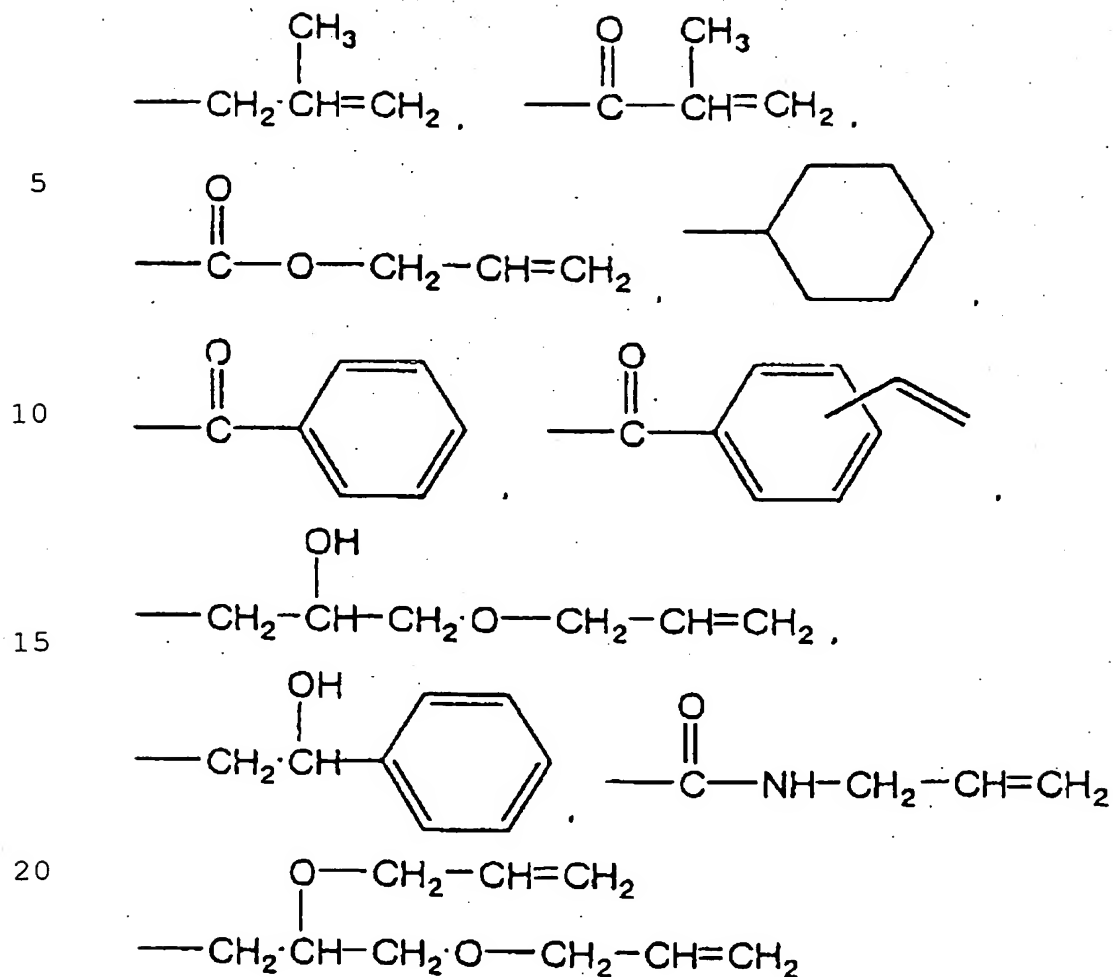
[0037]

From the viewpoint that the curing product obtained can have higher heat resistance, the group R^3 in the above general formula (III) is preferably a univalent organic group
 15 containing 1 to 20 carbon atoms, more preferably a univalent organic group containing 1 to 10 carbon atoms, still more preferably a univalent organic group containing 1 to 4 carbon atoms. As preferred examples of R^3 , there may be mentioned methyl, ethyl, propyl, butyl, phenyl, benzyl, phenethyl, vinyl,
 20 allyl, glycidyl,

[0038]

[Chemical 11]





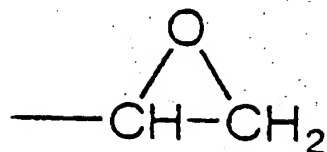
, and the like.

25 [0039]

From the viewpoint that the curing product to be obtained can be improved in adhesion to various materials, it is preferable that at least one of the three R³ groups in the above general formula (III) is preferably a univalent organic group containing 1 to 50 carbon atoms and containing one or more epoxy groups, more preferably a univalent organic group containing 1 to 50 carbon atoms and containing one or more epoxy groups represented by the formula.

[0040]

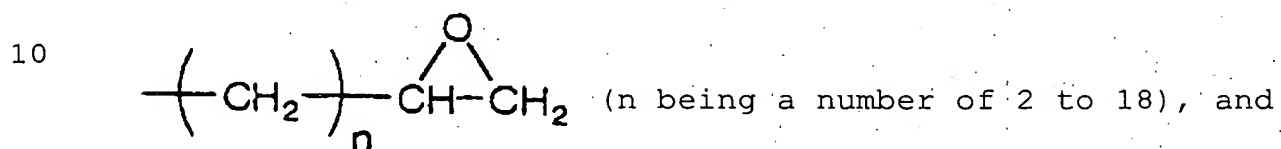
35 [Chemical 12]



5 As these preferable R^3 , there may be mentioned, for example, glycidyl,

[0041]

[Chemical 13]



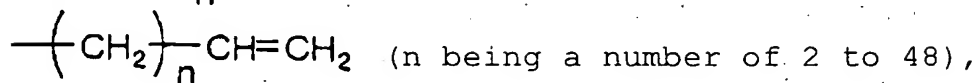
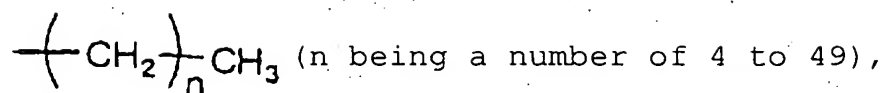
20 [0042]

From the viewpoint that the curing product to be obtained can be improved in chemical heat resistance, the R^3 group in the above general formula (III) is preferably a univalent organic group containing 1 to 50 carbon atoms and containing no or at most two oxygen atoms and comprising C, H, N and/or O, as exclusive constituent elements, and still more preferably a univalent hydrocarbon group containing 1 to 50 carbon atoms. As preferred examples of such R^3 , there may be mentioned methyl, ethyl, propyl, butyl, phenyl, benzyl, phenethyl, vinyl, allyl, glycidyl,

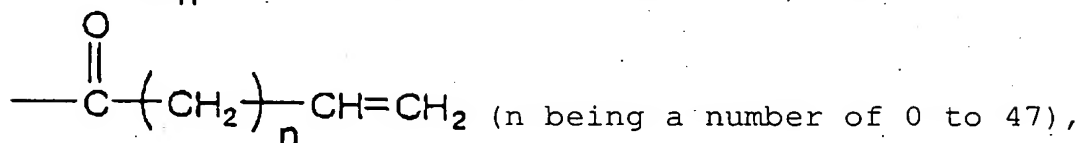
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[0043]

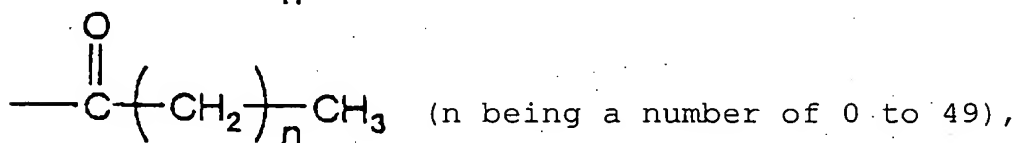
[Chemical 14]



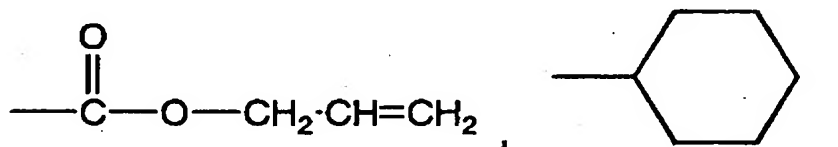
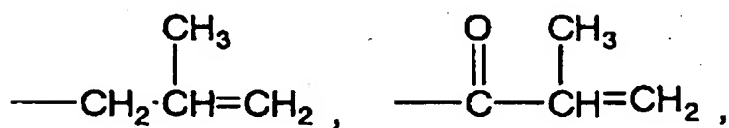
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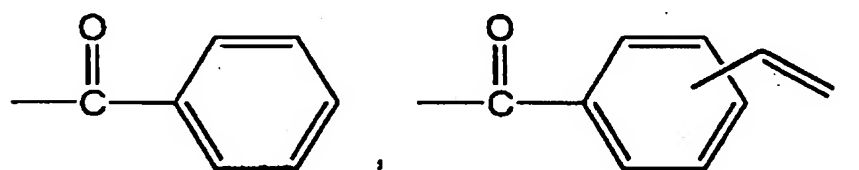
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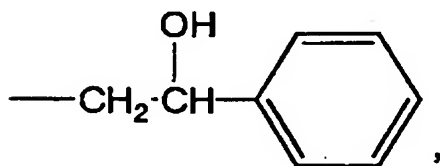
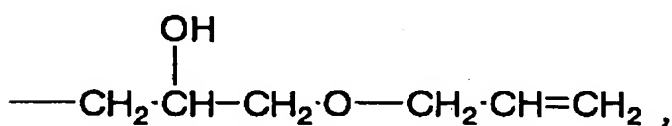
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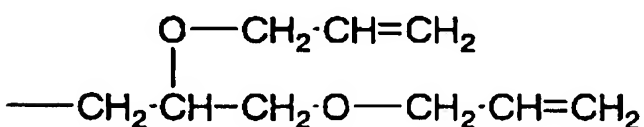
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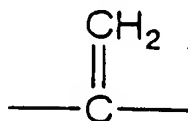
35 and the like.

[0044]

From the better reactivity viewpoint, at least one of the three R^3 groups in the above general formula (III) is preferably a univalent organic group containing 1 to 50 carbon atoms and containing one or more groups represented by the formula:

[0045]

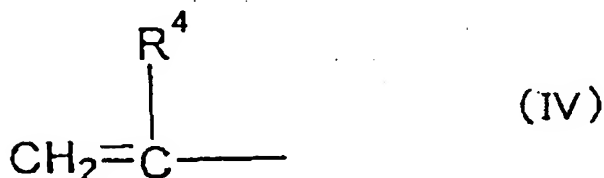
[Chemical 15]



More preferably, at least one of the three R^3 groups is a univalent organic group containing 1 to 50 carbon atoms and containing one or more groups represented by the following general formula (IV):

[0046]

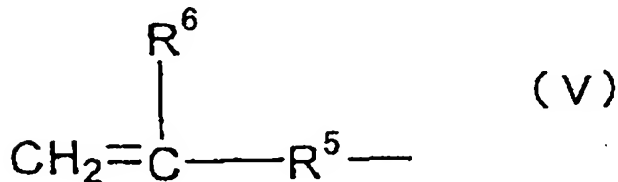
[Chemical 16]



wherein R^4 represents a hydrogen atom or a methyl group. Still more preferably, at least two of the three R^3 groups are organic compounds each represented by the following general formula (V):

[0047]

[Chemical 17]



wherein R^5 represents a direct bond or a bivalent organic group containing 1 to 48 carbon atoms and R^6 represents a hydrogen atom or a methyl group (the plurality each of R^5 and R^6 groups

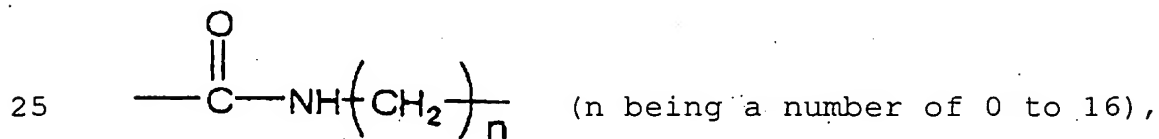
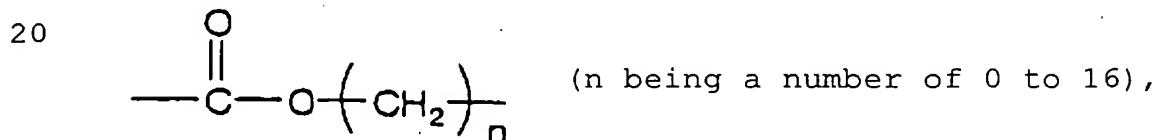
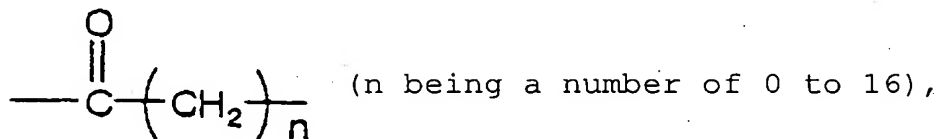
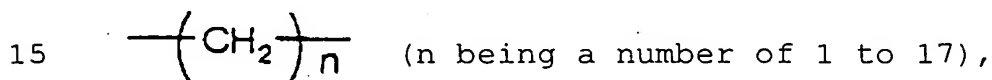
may be the same or different).

[0048]

From the viewpoint that the curing product obtained can be higher in heat resistance, R^5 in the above general formula (V), which may be a direct bond or a bivalent organic group containing 1 to 48 carbon atoms, is preferably a direct bond or a bivalent organic group containing 1 to 20 carbon atoms, more preferably a direct bond or a bivalent organic group containing 1 to 10 carbon atoms, still more preferably a direct bond or a bivalent organic group containing 1 to 4 carbon atoms. As examples of R^5 in such preferred cases, there may be mentioned

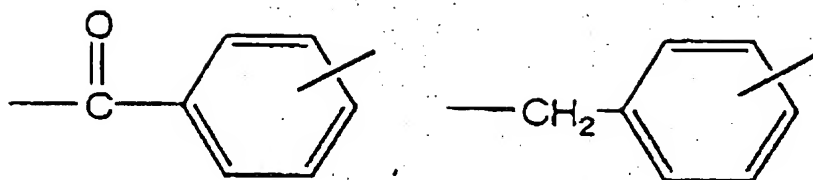
[0049]

[Chemical 18]

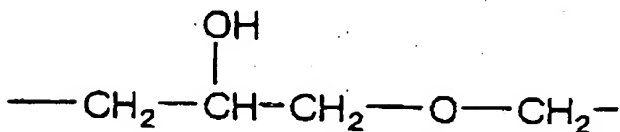


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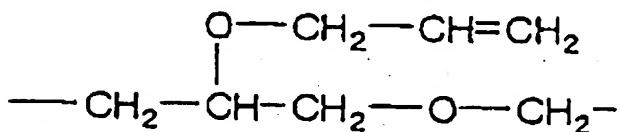
35



5



10



and the like.

[0050]

15

From the viewpoint that the curing product obtained can be improved in chemical heat stability, R^5 in the above general formula (V) is preferably a direct bond or a bivalent organic group containing 1 to 48 carbon atoms and containing not more than two oxygen atoms and containing C, H and/or O, as exclusive constituent elements, more preferably a direct bond or a bivalent hydrocarbon group containing 1 to 48 carbon atoms. As

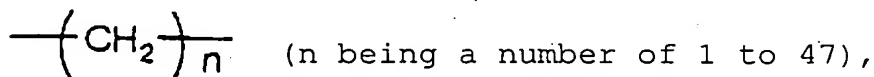
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examples of R^5 in such preferred cases, there may be mentioned

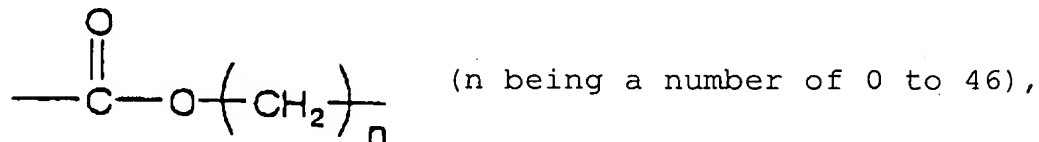
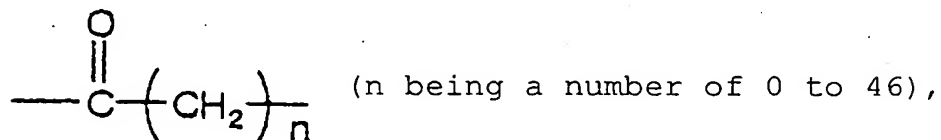
[0051]

[Chemical 19]

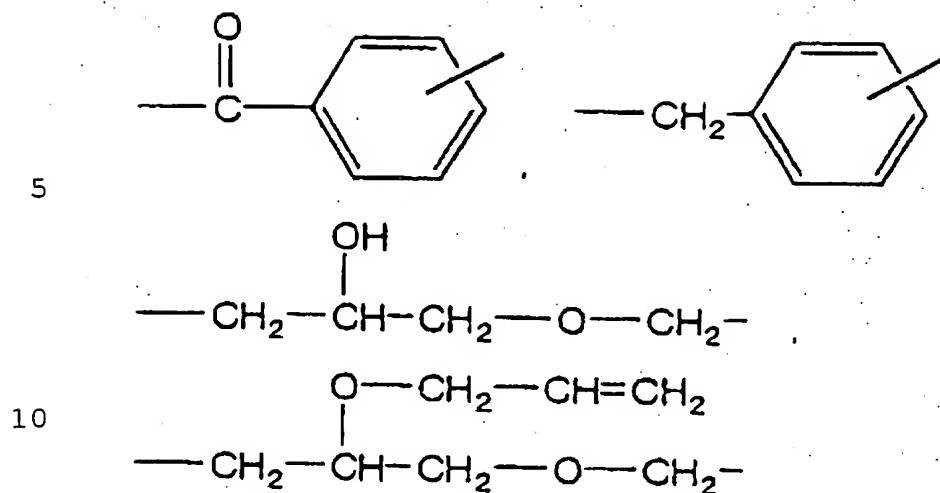
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30



35



and the like.

[0052]

15 From the better reactivity viewpoint, R^6 in the above general formula (V), which may be a hydrogen atom or a methyl group, is preferably a hydrogen atom.

[0053]

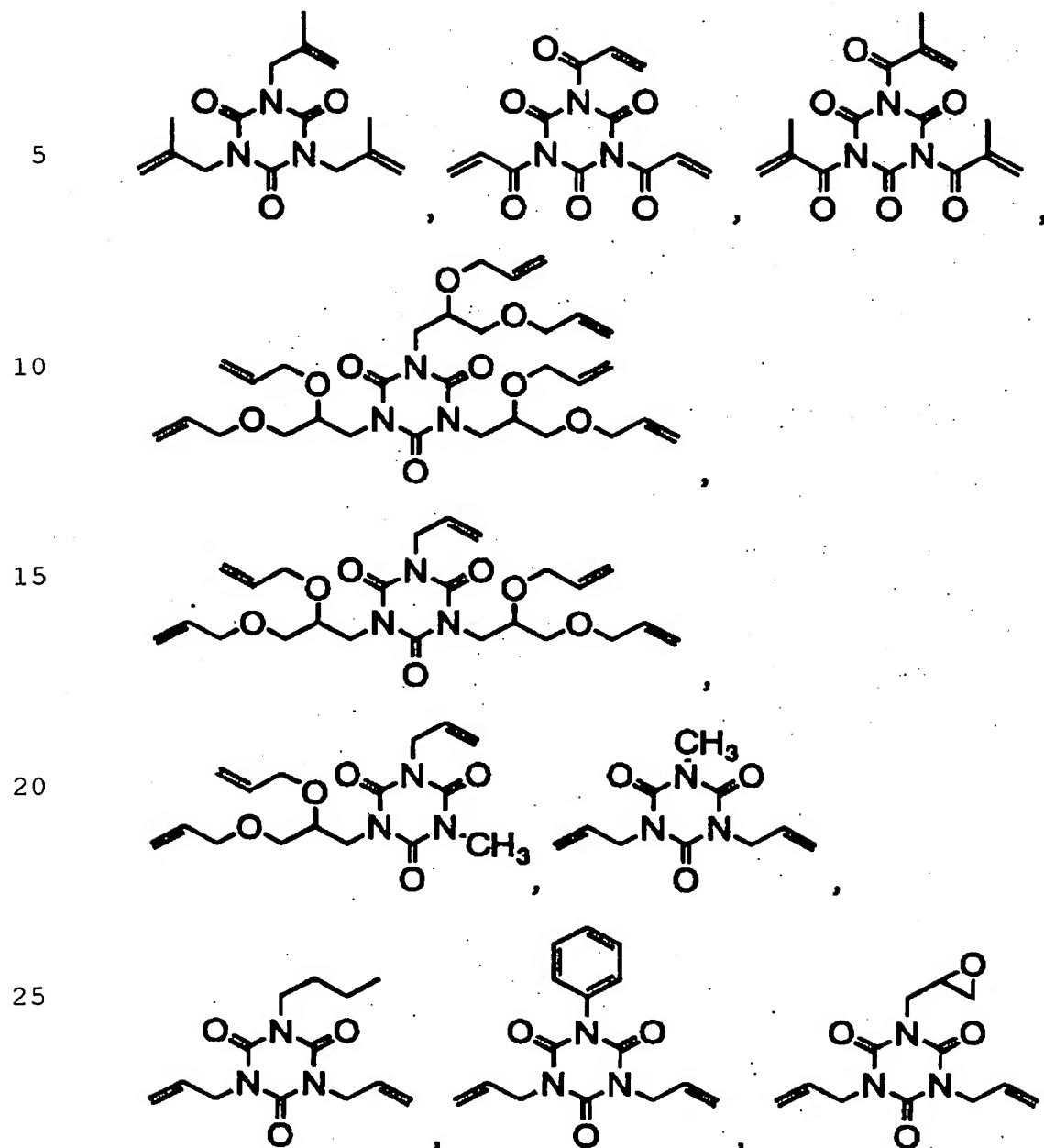
20 Even in the preferred examples of the organic compound of general formula (III) as mentioned above, however, it is necessary that there are at least two carbon-carbon double bonds reactive with a SiH group in each molecule. From the viewpoint of the possibility of further improvements in heat resistance, the compound of general formula (III) is more preferably an
25 organic compound containing three or more carbon-carbon double bonds reactive with a SiH group in each molecule.

[0054]

As preferred specific examples of the organic compound represented by the general formula (III), there may be mentioned
30 triallyl isocyanurate,

[0055]

[Chemical 20]



30 and the like.

The component (A) may be used alone, or as a mixture of two or more species of these.

Component (B)

35 An explanation is now made of the component (B), a compound

containing a SiH group.

[0056]

The component (B) of the invention is a compound containing at least two SiH groups in each molecule.

5 [0057]

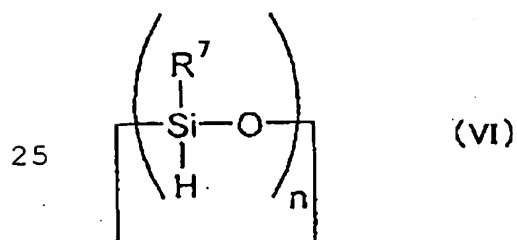
The component (B) may be any compound containing at least two SiH groups in each molecule, without any other restriction. Thus, for example, those compounds which are described in International Laid-open Patent Application WO 96/15194 and have at least two SiH groups in each molecule and the like can be used.

[0058]

From the availability viewpoint, linear and/or cyclic organopolysiloxanes having at least two SiH groups in each molecule are preferred, and, from the viewpoint of compatibility with the component (A), those cyclic organopolysiloxanes having at least two SiH groups in each molecule which are represented by the following general formula (VI):

20 [0059]

[Chemical 21]



wherein R^7 represents an organic group containing 1 to 6 carbon atoms and n represents a number of 3 to 10, are more preferred.

30 [0060]

The substituent R^7 in the compounds represented by the general formula (VI) is preferably a substituent containing C, H and/or O, more preferably a hydrocarbon group, still more preferably a methyl group.

35 [0061]

From the ready availability viewpoint, 1,3,5,7-tetramethylcyclotetrasiloxane is preferred as the compound represented by the general formula (VI).

[0062]

5 The molecular weight of the component (B) is not particularly restricted but may be optionally selected. From the viewpoint that the fluidity can be manifested more easily, low molecular weights are preferred. In this case, the lower limit to the molecular weight is preferably 50, and the upper
10 limit thereto is preferably 100,000, more preferably 1,000, still more preferably 700.

[0063]

The component (B) to be used may contain one single species or a mixture of two or more species.

15 [0064]

Further, from the viewpoint of good compatibility with the component (A) and from the viewpoint that the component (B) is low in volatility and the problem of outgassing from the resulting composition hardly occurs, those compounds which can
20 be obtained by subjecting an organic compound (α) having one or more carbon-carbon double bonds reactive with a SiH group in each molecule and a linear and/or cyclic polyorganosiloxane (β) having at least two SiH groups in each molecule to hydrosilylation reaction are preferred as the component (B).

25

(Component (α))

Usable here as the component (α) are organic compounds (α 1) which have at least two carbon-carbon double bonds reactive with a SiH group in each molecule and are to serve as the
30 component (A) mentioned above. The use of such component (α 1) tends to result in a high crosslink density and high mechanical strength of the curing product.

[0065]

In addition, as the component (α), compounds (α 2) having
35 one carbon-carbon double bond reactive with a SiH group in each

molecule can also be used. The use of the component (α_2) tends to render the curing product low in elasticity.

(Component (α_2))

The component (α_2) is not particularly restricted but may
 5 be any organic compound having one carbon-carbon double bond reactive with a SiH group in each molecule. From the viewpoint of better compatibility between the component (B) and the component (A), however, a compound other than a polysiloxane-organic block copolymer, a polysiloxane-organic
 10 graft copolymer or a like siloxane unit (Si-O-Si)-containing one is preferred, and a compound containing C, H, N, O, S and halogen, as exclusive constituent elements, is preferred.

[0066]

The sites of occurrence of the carbon-carbon double bonds
 15 reactive with a SiH group in component (α_2) are not particularly restricted but may be anywhere within the molecule.

[0067]

The component (α_2) compound may be classified as a polymer type compound or as a monomer type compound.

20 [0068]

Usable as the polymer type compound, there may be mentioned, for example, polysiloxane type, polyether type, polyester type, polyarylate type, polycarbonate type, saturated hydrocarbon type, unsaturated hydrocarbon type,
 25 polyacrylic ester type, polyamide type, phenol-formaldehyde type (phenol resin type), polyimide type compounds, etc.

[0069]

As the monomer type compound, there may be mentioned, for example, phenol type, bisphenol type, benzene, naphthalene or
 30 like aromatic hydrocarbon type compounds; aliphatic (e.g. chain, alicyclic, etc.) hydrocarbon type compounds; heterocyclic type compounds; silicon compounds; mixtures of these; etc.

[0070]

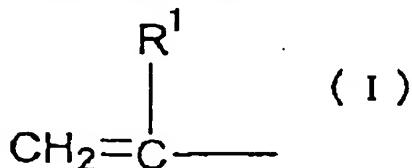
The carbon-carbon double bonds reactive with a SiH group
 35 in the component (α_2) are not particularly restricted but, from

the reactivity viewpoint, those groups represented by the following general formula (I):

[0071]

[Chemical 22]

5

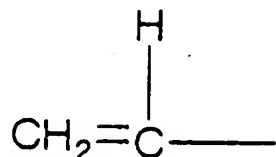


wherein R^1 represents a hydrogen atom or a methyl group, are preferred. From the material availability viewpoint, the group represented by

[0072]

[Chemical 23]

15



is particularly preferred.

[0073]

20

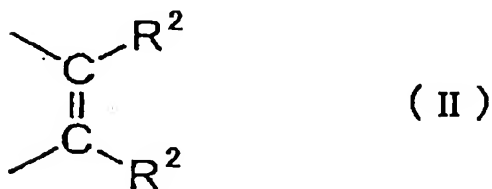
Furthermore, preferred as the carbon-carbon double bonds reactive with a SiH group in the component ($\alpha 2$) are alicyclic groups having a partial structure represented by the following general formula (II):

[0074]

25

[Chemical 24]

30

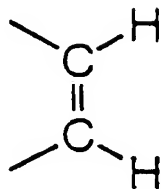


35

wherein each R^2 represents a hydrogen atom or a methyl group, in the ring structure, since high heat resistance can be given to curing products. From the material availability viewpoint, alicyclic groups having a partial structure represented by the following formula

[0075]

[Chemical 25]

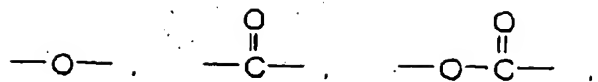


in the ring structure are preferred.

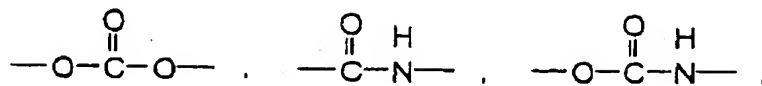
The carbon-carbon double bonds reactive with a SiH group each may be directly bonded to the skeletal portion of the component (α 2) or covalently bonded thereto via a bivalent or polyvalent substituent. The above bivalent or polyvalent substituent is not particularly restricted but is preferably a substituent containing 0 to 10 carbon atoms. From the viewpoint of better compatibility between the component (B) and the component (A), however, a substituent containing C, H, N, O, S and halogen, as exclusive constituent elements is preferred. Examples of the above substituent are:

[0076]

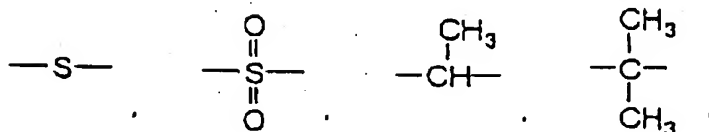
[Chemical 26]



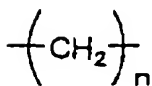
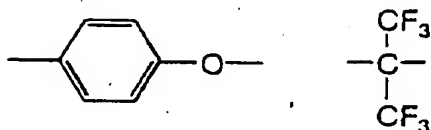
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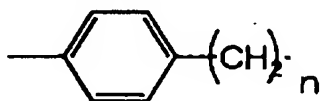
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15



20 (n representing a number of 1 to 10),



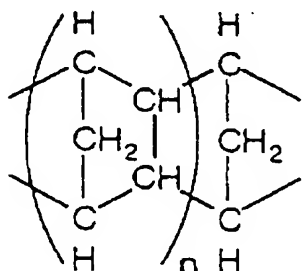
25

(n representing a number of 0 to 4), and

[0077]

[Chemical 27]

30



35

(n representing a number of 0 to 4), etc. Two or more of the bivalent or polyvalent substituents recited above may be

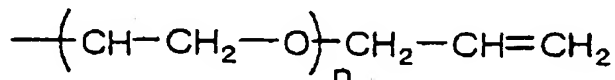
covalently bonded together to form one bivalent or polyvalent substituent.

[0078]

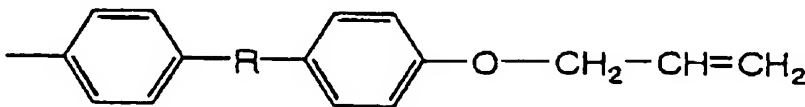
As examples of the group covalently bonded to the skeletal portion, there may be mentioned vinyl, allyl, methallyl, acryl, methacryl, 2-hydroxy-3-(allyloxy)propyl, 2-allylphenyl, 3-allylphenyl, 4-allylphenyl, 2-(allyloxy)phenyl, 3-(allyloxy)phenyl, 4-(allyloxy)phenyl, 2-(allyloxy)ethyl, 2,2-bis(allyloxymethyl)butyl, 3-allyloxy-2,2-bis(allyloxymethyl)propyl,

[0079]

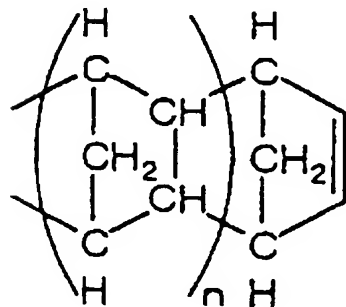
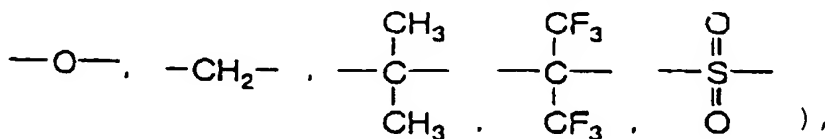
[Chemical 28]



(n representing a number satisfying the relation $5 \geq n \geq 2$),



(R representing a bivalent group selected from among



(n representing a number of 0 to 4), and the like.

[0080]

As specific examples of the component (α_2), there may be

mentioned linear aliphatic hydrocarbon compounds such as propene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 1-dodecene, 1-undecene, Idemitsu Petrochemical's Linealene, 4,4-dimethyl-1-pentene, 5 2-methyl-1-hexene, 2,3,3-trimethyl-1-butene and 2,4,4-trimethyl-1-pentene; alicyclic hydrocarbon compounds such as cyclohexene, methylcyclohexene, methylenecyclohexane, norbornylene, ethylidenecyclohexane, vinylcyclohexane, camphene, carene, α -pinene and β -pinene; aromatic hydrocarbon 10 compounds such as styrene, α -methylstyrene, indene, phenylacetylene, 4-ethynyltoluene, allylbenzene and 4-phenyl-1-butene; allyl ethers such as alkyl allyl ethers and allyl phenyl ether; aliphatic compounds such as glycerol monoallyl ether, ethylene glycol monoallyl ether and 15 4-vinyl-1,3-dioxolan-2-one; aromatic compounds such as 1,2-dimethoxy-4-allylbenzene and o-allylphenol; substituted isocyanurates such as monoallyl dibenzyl isocyanurate and monoallyl diglycidyl isocyanurate; silicon compounds such as vinyltrimethylsilane, vinyltrimethoxysilane and 20 vinyltriphenylsilane; etc.

Furthermore, as the component (α_2), there may be mentioned polymers and oligomers having a vinyl group at one end, for example polyether resins such as polyethylene oxide allylated at one end and polypropylene oxide allylated at one 25 end; hydrocarbon resins such as polyisobutylene allylated at one end; acrylic resins such as poly(butyl acrylate) allylated at one end and poly(methyl methacrylate) allylated at one end; etc.

[0081]

30 The structure may be linear or branched. The molecular weight is not particularly restricted but may be at any of various levels. The molecular weight distribution is not particularly restricted, either, but may be diverse. From the viewpoint that the mixture viscosity tends to become low and 35 the moldability tends to become better, however, the molecular

weight distribution is preferably not more than 3, more preferably not more than 2, still more preferably not more than 1.5.

[0082]

5 The glass transition temperature of the component (α_2), if any, is not particularly restricted, either, but may be diverse. In view of the tendency for the curing product obtained to become tough, however, the glass transition temperature is preferably not higher than 100°C, more
10 preferably not higher than 50°C, still more preferably not higher than 0°C. As preferred examples of the resin, there may be mentioned poly(butyl acrylate) and the like. Conversely, in view of the tendency for the curing product obtained to become highly resistant to heat, the glass transition temperature is
15 preferably not lower than 100°C, more preferably not lower than 120°C, still more preferably not lower than 150°C, most preferably not lower than 170°C.

 The glass transition temperature can be determined by dynamic viscoelasticity measurement and expressed in terms of
20 the temperature at which a maximum $\tan\delta$ value is obtained.

[0083]

 In view of the tendency for the curing product obtained to become high in heat resistance, the component (α_2) is preferably a hydrocarbon compound. In this case, the lower
25 limit to the number of carbon atoms is 7, and a preferred upper limit thereto is 10.

[0084]

 The component (α_2) may have other reactive group. As the above reactive group, there may be mentioned the following
30 groups, for example: epoxy, amino, radical-polymerizable unsaturated group, carboxyl, isocyanate, hydroxyl, alkoxysilyl group and so on. In cases where the above component has the above functional group, the adhesive properties of the curable composition obtained tends to become higher and the
35 strength of the curing product obtained tends to become high.

From the higher adhesive properties viewpoint, an epoxy group is preferred among those functional groups. In view of the tendency for the resulting curing product to have a higher level of heat resistance, it is preferred that the above component
5 have not less than one reactive group mentioned above per molecule on average. As specific examples, there may be mentioned monoallyl diglycidyl isocyanurate, allyl glycidyl ether, allyloxyethyl methacrylate, allyloxyethyl acrylate, vinyltrimethoxysilane, etc.

10 [0085]

The components (α_2) may be used singly or in combination of a plurality thereof.

(Component (β))

15 The component (β) is a linear and/or cyclic polyorganosiloxane having at least two SiH groups in each molecule.

[0086]

Specifically, there may be mentioned:

20 [0087]

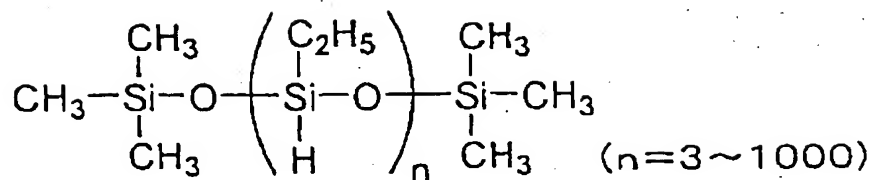
[Chemical 29]

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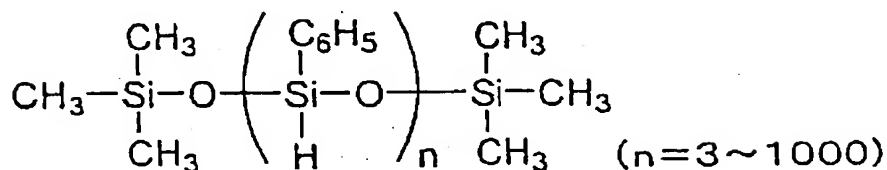
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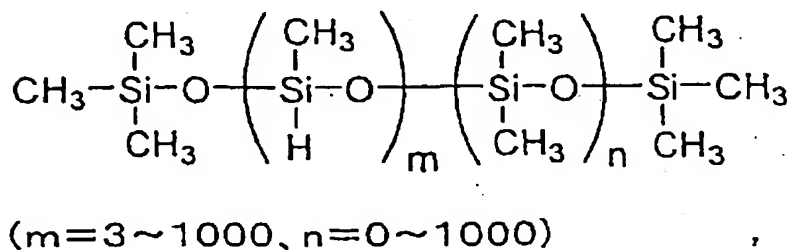
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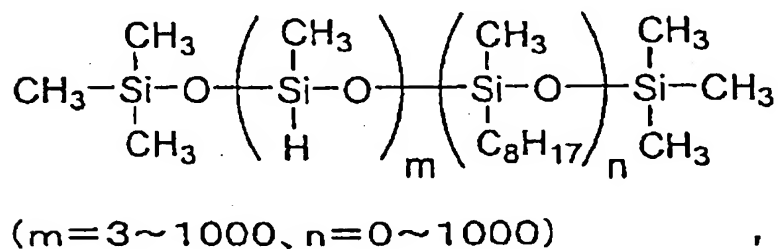
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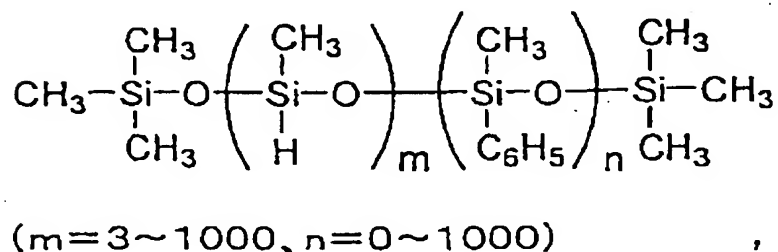
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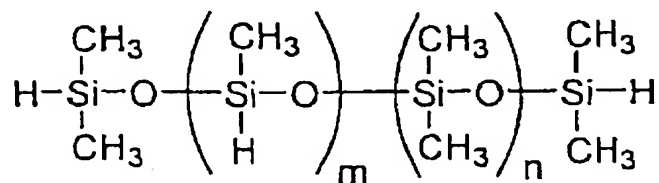
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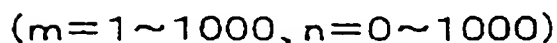
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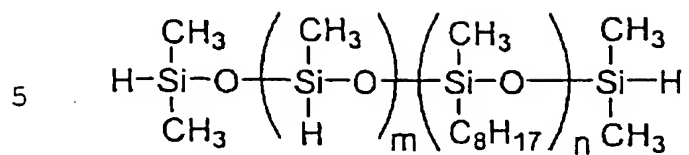


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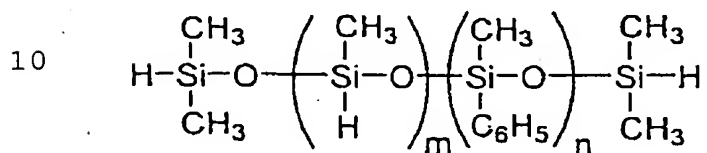


[0088]

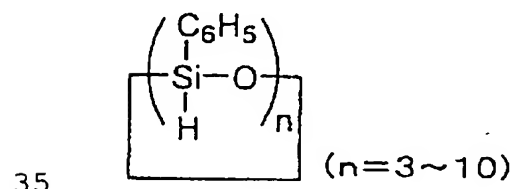
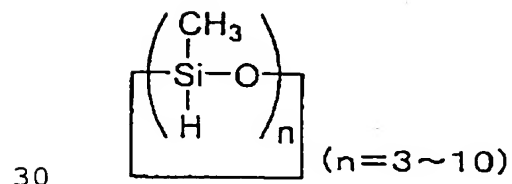
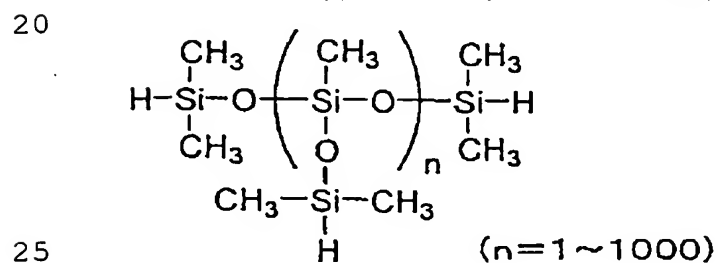
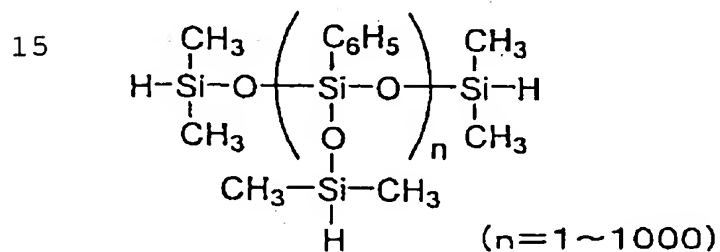
[Chemical 30]



(m=1~1000, n=0~1000)



(m=1~1000, n=0~1000)



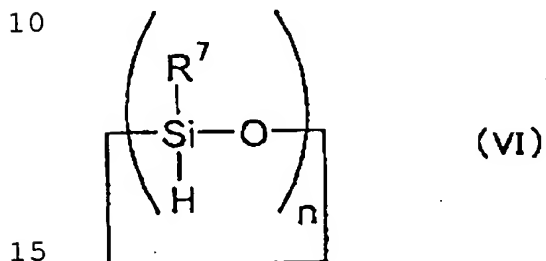
and the like.

[0089]

From the viewpoint that the compatibility with the component (α) tends to become better, those cyclic polyorganosiloxanes having at least three SiH groups in each molecule which are represented by the following general formula (VI):

[0090]

[Chemical 31]



wherein R^7 represents an organic group containing 1 to 6 carbon atoms and n represents a number of 3 to 10, are preferred.

[0091]

20 The substituent R^7 in the compounds represented by the general formula (VI) is preferably a substituent comprising C, H and/or O, more preferably a hydrocarbon group, still more preferably a methyl group.

[0092]

25 As the compounds represented by the general formula (IX), 1,3,5,7-tetramethylcyclotetrasiloxane is preferred from the ready availability viewpoint, etc.

[0093]

30 The component (β) may be used singly or as a mixture of two or more of them.

(Reaction between component (α) and component (β))

35 An explanation is now made of the hydrosilylation reaction between the component (α) and component (β), which is to be carried out in preparing a compound resulting from

hydrosilylation reaction between the component (α) and component (β) for use as the component (B) of the invention.

[0094]

The hydrosilylation reaction of the component (α) and component (β) may give a mixture of a plurality of compounds, including the component (B) of the invention in some instances. It is also possible to use the mixture as such in preparing the curable composition of the invention, without separating the component (B).

[0095]

The mixing ratio between the component (α) and component (β) in carrying out the hydrosilylation reaction between the component (α) and component (β) is not particularly restricted but, when the strength of the curing product resulting from hydrosilylation of the component (A) with the component (B) obtained is taken into consideration, a higher SiH group content in the component (B) is preferred, so that the ratio of the total number (X) of carbon-carbon double bonds reactive with a SiH group as occurring in the component (α) to be admixed and the total number (Y) of SiH groups in the component (β) to be admixed is preferably such that it is not less than 2 ($Y/X \geq 2$), more preferably such that it is not less than 3 ($Y/X \geq 3$). From the viewpoint that the compatibility of the component (B) with the component (A) tends to become better, it is preferred that the ratio satisfied the relation $10 \geq Y/X$, more preferably $5 \geq Y/X$.

[0096]

In subjecting the component (α) and component (β) to hydrosilylation reaction, an appropriate catalyst may be used. As the catalyst, there may be mentioned simple substance platinum, solid platinum supported on such a carrier as alumina, silica or carbon black, chloroplatinic acid, complexes of chloroplatinic acid with an alcohol, aldehyde, ketone, etc., platinum-olefin complexes (e.g. $\text{Pt}(\text{CH}_2=\text{CH}_2)_2(\text{PPh}_3)_2$, $\text{Pt}(\text{CH}_2=\text{CH}_2)_2\text{Cl}_2$), platinum-vinylsiloxane complexes (e.g. $\text{Pt}(\text{ViMe}_2\text{SiOSiMe}_2\text{Vi})_n$, $\text{Pt}[(\text{MeViSiO})_4]_m$), platinum-phosphine

complexes (e.g. $\text{Pt}(\text{PPh}_3)_4$, $\text{Pt}(\text{PBu}_3)_4$), platinum-phosphite complexes (e.g. $\text{Pt}[\text{P}(\text{OPh})_3]_4$, $\text{Pt}[\text{P}(\text{OBu})_3]_4$) (in the formulas, Me represents a methyl group, Bu a butyl group, Vi a vinyl group and Ph a phenyl group, and n and m each represents an integer),
 5 dicarbonyldichloroplatinum, Karstedt catalyst, platinum-hydrocarbon complexes described in Ashby's U. S. Patent No. 3,159,601 and 3,159,662, platinum-alcoholate catalysts described in Lamoreaux's U. S. Patent No. 3,220,972, and so on. Furthermore, platinum chloride-olefin complexes
 10 described in Modic's U. S. Patent No. 3,516,946 are also useful in the practice of the present invention.

[0097]

As catalysts other than platinum compounds, there may be mentioned, for example, $\text{RhCl}(\text{PPh})_3$, RhCl_3 , RhAl_2O_3 , RuCl_3 , IrCl_3 ,
 15 FeCl_3 , AlCl_3 , $\text{PdCl}_2 \cdot 2\text{H}_2\text{O}$, NiCl_2 , TiCl_4 , etc.

[0098]

Among these, chloroplatinic acid, platinum-olefin complexes, platinum-vinylsiloxane complexes and the like are preferred from the catalytic activity viewpoint. These
 20 catalysts may be used singly or in combination of two or more.

[0099]

The level of addition of the catalyst is not particularly restricted but, for attaining a sufficient level of curability and reducing the cost of the curable composition to a relatively
 25 low level, the lower limit to the addition level is preferably 10^{-8} mole, more preferably 10^{-6} mole, per mole of the SiH group in the component (β), and the upper limit to the addition level is preferably 10^{-1} mole, more preferably 10^{-2} mole, per mole of the SiH group in the component (β).

30 [0100]

A promoter may be used in combination with the above catalyst. As the promoter, there may be mentioned, for example, phosphorus compounds such as triphenylphosphine; 1,2-diester compounds such as dimethyl maleate; acetylene alcohol compounds
 35 such as 2-hydroxy-2-methyl-1-butyne; sulfur compounds

inclusive of simple substance sulfur; amine compounds such as triethylamine; etc. The level of addition of the promoter is not particularly restricted but the lower limit to the addition level is preferably 10^{-2} mole, more preferably 10^{-1} mole, and the upper limit is 10^2 mole, more preferably 10 moles, per mole of said hydrosilyl catalyst.

[0101]

As for the method of mixing up the component (α), component (β) and catalyst in carrying out the reaction, various methods may be employed. Preferred is, however, the method comprising mixing the component (α) with the catalyst and mixing the resulting mixture with the component (β). The method comprising mixing the catalyst with a mixture of the component (α) and component (β) makes it difficult to control the reaction. When the method comprising mixing the component (α) with a mixture of the component (β) and catalyst is employed, the component (β), which is reactive with moisture possibly contained therein in the presence of the catalyst, may cause denaturation in some instances.

[0102]

The reaction temperature may be selected at various levels. In the instance case, the lower limit to the reaction temperature is preferably 30°C , more preferably 50°C , and the upper limit to the reaction temperature is preferably 200°C , more preferably 150°C . At lower reaction temperatures, the reaction time for the reaction to proceed to a satisfactory extent becomes long whereas higher reaction temperatures are not practical. The reaction may be carried out at a constant temperature, or the temperature may be varied stepwise or continuously according to need.

[0103]

The reaction time and the pressure during reaction may also be selected at various levels according to need.

[0104]

A solvent may be used in the hydrosilylation reaction.

The solvent to be used is not particularly restricted but any of those which will not inhibit the hydrosilylation reaction. Specifically, hydrocarbon solvents such as benzene, toluene, hexane and heptane; ether solvents such as tetrahydrofuran, 1,4-dioxane, 1,3-dioxolane and diethyl ether; ketone solvents such as acetone and methyl ethyl ketone; and halogenated solvents such as chloroform, methylene chloride and 1,2-dichloroethane can be used satisfactorily. Such solvents can also be used in the form of a mixed solvent composed of two or more species. Preferred as the solvent are toluene, tetrahydrofuran, 1,3-dioxolane and chloroform. The amount of the solvent to be used can be adequately selected.

[0105]

Further, various additives may be used for the purpose of controlling the reactivity, etc.

[0106]

It is also possible, after reacting the component (α) with the component (β), to remove the solvent, unreacted component (α) and/or unreacted component (β). When these volatile components are removed, the component (B) obtained becomes free of such volatile components and, therefore, the problems of void formation and cracking due to volatile matter evaporation in the step of curing with the component (A) will hardly occur. The method of removal includes, for example, volatile matter removal under reduced pressure and, further, treatment with active carbon, aluminum silicate, silica gel or the like. In the case of volatile matter removal under reduced pressure, the treatment is preferably carried out at low temperatures. In this case, the upper limit to the temperature is preferably 100°C, more preferably 60°C. High temperature treatment tends to be accompanied by changes in properties, for example viscosity increase.

[0107]

As examples of the component (B), namely a reaction product from the components (α) and (β), there may be mentioned

the bisphenol A diallyl ether-1,3,5,7-tetramethylcyclotetrasiloxane reaction product, vinylcyclohexene-1,3,5,7-tetramethylcyclotetrasiloxane reaction product,

5 divinylbenzene-1,3,5,7-tetramethylcyclotetrasiloxane reaction product, dicyclopentadiene-1,3,5,7-tetramethylcyclotetrasiloxane reaction product, triallyl isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction

10 product, diallyl monoglycidyl isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction product, allyl glycidyl ether-1,3,5,7-tetramethylcyclotetrasiloxane reaction product, (α)-methylstyrene-1,3,5,7-tetramethylcyclotetrasiloxane

15 reaction product, monoallyl diglycidyl isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction product, etc. From the viewpoint of the heat resistance and light resistance, the triallyl isocyanurate-1,3,5,7-tetramethylcyclotetrasiloxane reaction

20 product is preferred.

[0108]

The component (C), a hydrosilylation catalyst is now explained.

[0109]

25 The hydrosilylation catalyst is not particularly restricted but may be any one having catalytic activity in the hydrosilylation reaction. Thus, there may be mentioned, for example, simple substance platinum, solid platinum supported on such a carrier as alumina, silica or carbon black,

30 chloroplatinic acid, complexes of chloroplatinic acid with an alcohol, aldehyde, ketone, etc., platinum-olefin complexes (e.g. $\text{Pt}(\text{CH}_2=\text{CH}_2)_2(\text{PPh}_3)_2$, $\text{Pt}(\text{CH}_2=\text{CH}_2)_2\text{Cl}_2$), platinum-vinylsiloxane complexes (e.g. $\text{Pt}(\text{ViMe}_2\text{SiOSiMe}_2\text{Vi})_n$, $\text{Pt}[(\text{MeViSiO})_4]_m$), platinum-phosphine complexes (e.g. $\text{Pt}(\text{PPh}_3)_4$,

35 $\text{Pt}(\text{PBu}_3)_4$), platinum-phosphite complexes (e.g. $\text{Pt}[\text{P}(\text{OPh})_3]_4$,

Pt[P(OBu)₃]₄) (in the formulas, Me represents a methyl group, Bu a butyl group, Vi a vinyl group and Ph a phenyl group, and n and m each represents an integer), dicarbonyldichloroplatinum, Karstedt catalyst, platinum-hydrocarbon complexes described in Ashby's U. S. Patents No. 3,159,601 and 3,159,662, platinum-alcoholate catalysts described in Lamoreaux's U. S. Patent No. 3,220,972, and so on. Furthermore, platinum chloride-olefin complexes described in Modic's U. S. Patent No. 3,516,946 are also useful in the practice of the present invention.

[0110]

As catalysts other than platinum compounds, there may be mentioned, for example, RhCl(PPh)₃, RhCl₃, RhAl₂O₃, RuCl₃, IrCl₃, FeCl₃, AlCl₃, PdCl₂·2H₂O, NiCl₂, TiCl₄, etc.

[0111]

Among these, chloroplatinic acid, platinum-olefin complexes, platinum-vinylsiloxane complexes and the like are preferred from the catalytic activity viewpoint. The above catalysts may be used singly or in combination of two or more.

[0112]

The level of addition of the catalyst is not particularly restricted but, for attaining a sufficient level of curability and reducing the cost of the curable composition to a relatively low level, the lower limit to the addition level is preferably 10⁻⁸ mole, more preferably 10⁻⁶ mole, per mole of the SiH group in the component (B), and the upper limit to the addition level is preferably 10⁻¹ mole, more preferably 10⁻² mole, per mole of the SiH group in the component (B).

[0113]

A promoter may be used in combination with the above catalyst. As the promoter, there may be mentioned, for example, phosphorus compounds such as triphenylphosphine; 1,2-diester compounds such as dimethyl maleate; acetylene alcohol compounds such as 2-hydroxy-2-methyl-1-butyne; sulfur compounds inclusive of simple substance sulfur; amine compounds such as

triethylamine; etc. The level of addition of the promoter is not particularly restricted but the lower limit to the addition level is preferably 10^{-2} mole, more preferably 10^{-1} mole, and the upper limit is 10^2 mole, more preferably 10 moles, per mole
5 of said hydrosilylation catalyst.

[0114]

The component (D), a silane coupling agent, is now explained.

[0115]

10 The silane coupling agent is not particularly restricted but may be any of those compounds which have, within the molecule, at least one functional group reactive with an organic group and a hydrolyzable silyl group.

[0116]

15 The functional group reactive with an organic group is not particularly restricted but at least one functional group selected from among epoxy, methacryl, acryl, isocyanate, isocyanurate, vinyl and carbamate groups is preferred from the handleability viewpoint. From the viewpoint of curability and
20 adhesive properties, epoxy, methacryl and acryl groups are more preferred. As the hydrolyzable silyl group, from the handleability viewpoint, preferred are alkoxysilyl groups and, from the reactivity viewpoint, methoxysilyl and ethoxysilyl groups are particularly preferred.

25 [0117]

As preferred examples of the silane coupling agent, there may be mentioned epoxy group-containing alkoxysilanes such as 3-glycidoxypropyltrimethoxysilane,
3-glycidoxypropyltriethoxysilane,
30 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane and 2-(3,4-epoxycyclohexyl)ethyltriethoxysilane; and methacryl or acryl group-containing alkoxysilanes such as 3-methacryloxypropyltrimethoxysilane,
3-methacryloxypropyltriethoxysilane,
35 3-acryloxypropyltrimethoxysilane,

3-acryloxypropyltriethoxysilane,
methacryloxymethyltrimethoxysilane,
methacryloxymethyltriethoxysilane,
acryloxymethyltrimethoxysilane and
5 acryloxymethyltriethoxysilane.

[0118]

The level of addition of the silane coupling agent can be selected at various levels. However, the lower limit to the addition level is preferably 0.1 part by weight, more preferably
10 0.5 part by weight, and the upper limit to the addition level is preferably 50 parts by weight, more preferably 25 parts by weight, per 100 parts by weight of [component (A) + component (B)]. At lower addition levels, the adhesive property-improving effect is hardly exerted and, at higher
15 addition levels, the physical properties of the curing product may possibly be adversely affected.

[0119]

The epoxy group-containing compound includes various epoxy resins, for example, novolak phenol-based epoxy resins,
20 biphenyl-based epoxy resins, dicyclopentadiene-based epoxy resins, bisphenol F diglycidyl ether, bisphenol A diglycidyl ether, 2,2'-bis(4-glycidyloxycyclohexyl)propane, 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexanecarboxylate, vinylcyclohexene dioxide,
25 2-(3,4-epoxycyclohexyl)-5,5-spiro(3,4-epoxycyclohexane)-1,3-dioxane, bis(3,4-epoxycyclohexyl) adipate, bisglycidyl 1,2-cyclopropanedicarboxylate, triglycidyl isocyanurate, monoallyl diglycidyl isocyanurate, diallyl monoglycidyl isocyanurate, and like epoxy resins as cured with an aliphatic
30 acid anhydride such as hexahydrophthalic anhydride, methylhexahydrophthalic anhydride, trialkyltetrahydrophthalic anhydrides or hydrogenated methylnadic anhydride, and the like. The epoxy resins, if necessarily a curing agent, may respectively be used singly or
35 in the form of a combination of a plurality of species.

[0120]

The level of addition of the epoxy group-containing compound can be selected at various levels. However, the lower limit to the addition level is preferably 0.1 part by weight, more preferably 0.5 part by weight, and the upper limit to the addition level is preferably 50 parts by weight, more preferably 25 parts by weight, per 100 parts by weight of [component (A) + component (B)]. At lower addition levels, the adhesive property-improving effect is hardly exerted and, at higher addition levels, the physical properties of the curing product may possibly be adversely affected.

[0121]

The component (E), a borate ester, is now explained. A borate ester is used as a silanol condensation catalyst, which can improve the adhesive properties and/or stabilization.

As the borate esters mentioned, a compound represented by the following general formula (VII) or (VIII):

[0122]

[Chemical 32]

20 $B(OR^8)_3$ (VII)

[0123]

[Chemical 33]

$B(OCOR^8)_3$ (VIII)

in each formula, R^8 represents an organic group containing 1 to 48 carbon atoms, may be suitably used.

As specific (E) components, preferably used may be tri-2-ethylhexyl borate, normaltrioctadecyl borate, trinormaloctyl borate, triphenyl borate, trimethylene borate, tris(trimethylsilyl) borate, trinormalbutyl borate, tri-sec-butyl borate, tri-tert-butyl borate, triisopropyl borate, trinormalpropyl borate, triallyl borate, triethyl borate, trimethyl borate, boron methoxyethoxide and the like. The (E) components may be used singly or in combination of two or more. The mixing may be carried out preliminary, or during cured product-production.

[0124]

From the availability viewpoint, trimethyl borate, triethyl borate, and trinormalbutyl borate are more preferred, and trimethyl borate is still more preferred.

5 [0125]

From the viewpoint of the possibility of volatility suppression in the step of curing, trinormaloctadecyl borate, trinormaloctyl borate, triphenyl borate, trimethylene borate, tris(trimethylsilyl) borate, trinormalbutyl borate, 10 tri-sec-butyl borate, tri-tert-butyl borate, triisopropyl borate, trinormalpropyl borate, triallyl borate, and boron methoxyethoxide are preferred, and normaltrioctadecyl borate, tri-tert-butyl borate, triphenyl borate, and trinormalbutyl borate are still more preferred.

15 [0126]

From the volatility suppression and/or workability viewpoint, trinormalbutyl borate, triisopropyl borate, and trinormalpropyl borate are more preferred, and trinormalbutyl borate is still more preferred.

20 [0127]

From the viewpoint of low degrees of coloration at high temperatures, triethyl borate is more preferred, and trimethyl borate is still more preferred.

[0128]

25 The level of addition of the silanol condensation catalyst can be selected at various levels. However, the lower limit to the addition level is preferably 0.1 part by weight, more preferably 1 part by weight, and the upper limit to the addition level is preferably 50 parts by weight, more preferably 30 30 parts by weight, per 100 parts by weight of the coupling agent. At lower addition levels, the adhesive property-improving effect is hardly exerted and, at higher addition levels, the physical properties of the curing product may possibly be adversely affected.

35 [0129]

In the present invention, a silanol source compound may be used such that adhesive-property improving effect can be more enhanced, whereby further improvement of the adhesive properties and/or stabilization can be achieved. The silanol source compound herein refers to a compound containing one or more silanol groups or alkoxysilyl groups in one molecule. As the above silanol source compound, there may be mentioned, for example, silanol compounds such as triphenyl silanol and diphenyldihydroxysilane, alkoxysilanes such as diphenyldimethoxysilane, tetramethoxysilane and methyltrimethoxysilane, etc.

[0130]

When such a silanol source compound is used, the amount thereof to be used can be selected at various levels. However, the lower limit to the addition level is preferably 0.1 part by weight, more preferably 1 part by weight, and the upper limit to the addition level is preferably 50 parts by weight, more preferably 30 parts by weight, per 100 parts by weight of the coupling agent and/or the epoxy compound.

At lower addition levels, the adhesive property-improving effect is hardly exerted and, at higher addition levels, the physical properties of the curing product may possibly be adversely affected.

[0131]

The above silanol source compounds may be used singly or two or more of them may be used in combination.

[0132]

For modifying the characteristics of the curable composition of the invention, it is also possible to add thermosetting resins thereto. The above thermosetting resin is not particularly restricted, but there may be mentioned, for example, epoxy resins, cyanate ester resins, phenol resins, polyimide resins, urethane resins, bismaleimide resins, etc. Among them, epoxy resins are preferred from the viewpoint that they are excellent in the practical properties such as

adhesiveness.

[0133]

The epoxy resin includes, for example, novolak phenol-based epoxy resins, biphenyl-based epoxy resins, 5 dicyclopentadiene-based epoxy resins, bisphenol F diglycidyl ether, bisphenol A diglycidyl ether, 2,2'-bis(4-glycidyloxycyclohexyl)propane, 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexanecarboxylate, vinylcyclohexene dioxide, 10 2-(3,4-epoxycyclohexyl)-5,5-spiro(3,4-epoxycyclohexane)-1,3-dioxane, bis(3,4-epoxycyclohexyl) adipate, bisglycidyl 1,2-cyclopropanedicarboxylate, triglycidyl isocyanurate, monoallyl diglycidyl isocyanurate, diallyl monoglycidyl isocyanurate, and like epoxy resins as cured with an aliphatic 15 acid anhydride such as hexahydrophthalic anhydride, methylhexahydrophthalic anhydride, trialkyltetrahydrophthalic anhydrides or hydrogenated methyl nadic anhydride, and the like. The epoxy resin or a curing agent may respectively be used singly or in the form of 20 a combination of a plurality of species.

[0134]

The level of addition of the above thermosetting resin is not particularly restricted. However, the lower limit to the amount to be used is preferably 5% by weight, more preferably 25 10% by weight, and the upper limit to the amount to be used is preferably 50% by weight, more preferably 30% by weight, of the total weight of the curable composition. At lower addition levels, the desired effects, such as adhesive properties, can hardly be obtained and, at higher levels, the curing product 30 tends to become brittle.

[0135]

The thermosetting resin may comprise one single species or a combination of a plurality of species.

[0136]

35 A raw resin and/or a cured resin of the thermosetting resin

may be mixed with the component (A) and/or (B) by homogeneously dissolving therein, dispersing therein in the form of ground particles, dispersing therein in the form of a solution in a solvent, or the like. From the viewpoint that the resulting curing product readily becomes more transparent, the resin is preferably mixed with the component (A) and/or (B) by homogeneously dissolving therein. In this case, too, the thermosetting resin may be directly dissolved in the component (A) and/or (B) or homogeneously mixed therewith using a solvent or the like, or the solvent may be removed thereafter to attain a homogeneous dispersion state and/or mixture state.

[0137]

When the thermosetting resin is dispersed, the average particle diameter can be selected at various levels. Preferably, however, the lower limit to the average particle diameter is 10 nm, and the upper limit to the average particle diameter is preferably 10 μ m. The particle system may have a distribution and may be monodisperse or show a plurality of peak particle diameters. From the viewpoint that the curable composition tend to be low in viscosity and show better moldability, the coefficient of variation in particle diameter is preferably not more than 10%.

[0138]

For modifying the characteristics of the composition of the invention, it is also possible to incorporate thermoplastic resins thereto. The above thermoplastic resin is not particularly restricted, but for example, there may be mentioned acrylic resins, typically poly(methyl methacrylate)-based resins such as methyl methacrylate homopolymers, or random, block or graft copolymers of methyl methacrylate and another monomer, or other polymers (e.g. Optorez, product of Hitachi Chemical, etc.), and poly(butyl acrylate)-based resins such as butyl acrylate homopolymers, or random, block or graft polymers of butyl acrylate and another monomer, or other polymers; polycarbonate-type resins such as

polycarbonate resins containing bisphenol A, 3,3,5-trimethylcyclohexylidenebisphenol or the like as a monomer structure (e.g. APEC, product of Teijin, etc.); cycloolefin resins such as resins resulting from

5 homopolymerization or copolymerization of norbornene derivatives, vinyl monomers and so forth, resins resulting from ring-opening metathetic polymerization of norbornene derivatives, or hydrogenation products derived therefrom (e.g. APEL, product of Mitsui Chemical; ZEONOR and ZEONEX, products

10 of Zeon Corporation; ARTON, product of JSR; etc.); olefin-maleimide-based resins such as ethylene-maleimide copolymers (e.g. TI-PAS, product of Tosoh, etc.); polyester resins such as polyesters resulting from polycondensation of a bisphenol such as bisphenol A or

15 bis(4-(2-hydroxyethoxy)phenyl)fluorine and/or a diol such as diethylene glycol and a phthalic acid such as terephthalic acid or isophthalic acid and/or an aliphatic dicarboxylic acid (e.g. O-PET, product of Kanebo, etc.); polyethersulfone resins; polyarylate resins; poly(vinyl acetal) resins; polyethylene

20 resins; polypropylene resins; polystyrene resins; polyamide resins; silicone resins, fluororesins; rubber-like resins such as natural rubbers and EPDM; etc.

[0139]

The above thermoplastic resin may have a carbon-carbon

25 double bond(s) reactive with a SiH group and/or a SiH group(s) within the molecule. In view of the tendency for the resulting curing product to have an increased level of toughness, it is preferred that it has at least one carbon-carbon double bond reactive with a SiH group and/or at least one SiH group per

30 molecule on average.

[0140]

The above thermoplastic resin may have another or other crosslinking group or groups. As the above crosslinking groups, there may be mentioned epoxy, amino, radical-polymerizable

35 unsaturated groups, carboxyl, isocyanate, hydroxyl,

alkoxysilyl groups, etc. In view of the tendency for the resulting curing product to have an increased level of heat resistance, the occurrence of at least one crosslinking group per molecule on average is preferred.

5 [0141]

The molecular weight of the above thermoplastic resin is not particularly restricted but, from the viewpoint of better compatibility with the component (A) and/or (B), the number average molecular weight is preferably not more than 10,000, more preferably not more than 5,000. Conversely, from the viewpoint of the tendency for the resulting curing product to readily become tougher, the number average molecular weight is preferably not less than 10,000, more preferably not less than 100,000. The molecular weight distribution is not particularly restricted, either. In view of the tendency for the mixture to have a lower viscosity and better moldability, however, the molecular weight distribution is preferably not more than 3, more preferably not more than 2, still more preferably not more than 1.5.

20 [0142]

The level of incorporation of the thermoplastic resin is not particularly restricted but the lower limit to the amount to be used is preferably 5% by weight, more preferably 10% by weight, of the whole amount of the curable composition, and the upper limit to the amount to be used is preferably 50% by weight, more preferably 30% by weight, in the curable composition. When the addition level is lower, the curing product obtained tends to be brittle while higher addition levels tends to result in decreased heat resistance (elasticity at elevated temperatures).

30

[0143]

The thermoplastic resin can be used singly or in a combination of a plurality of species.

[0144]

35 The above thermoplastic resin may be mixed with the

component (A) and/or (B) by homogeneously dissolving therein, dispersing therein in the form of ground particles, dispersing therein in the form of a solution in a solvent, or the like. From the viewpoint that the resulting curing product readily becomes more transparent, the resin is preferably mixed with the component (A) and/or (B) by homogeneously dissolving therein. In this case, too, the thermoplastic resin may be directly dissolved in the component (A) and/or (B) or homogeneously mixed therewith using a solvent or the like, or the solvent may be removed thereafter to attain a homogeneous dispersion state and/or mixture state.

[0145]

When the thermoplastic resin is dispersed, the average particle diameter can be selected at various levels. Preferably, however, the lower limit to the average particle diameter is 10 nm, and the upper limit to the average particle diameter is preferably 10 μ m. The particle system may have a distribution and may be monodisperse or show a plurality of peak particle diameters. In view of the tendency for the curable composition to readily become low in viscosity and show better moldability, the coefficient of variation in particle diameter is preferably not more than 10%.

[0146]

A filler may be added to the curable composition of the invention.

[0147]

The above filler may be various ones, for example, silica type filler such as quartz, fumed silica, precipitated silica, silicic anhydride, fused silica, crystalline silica and ultrafine amorphous silica; inorganic fillers such as silicon nitride, silver powders, alumina, aluminum hydroxide, titanium oxide, glass fibers, carbon fibers, mica, carbon black, graphite, diatomaceous earth, terra alba, clay, talc, calcium carbonate, magnesium carbonate, barium sulfate and inorganic ballon; those fillers in common use in and/or proposed as

fillers for the conventional sealing member, such as an epoxy type filler; and the like.

[0148]

As the filler, those low in radioactivity are preferred
5 from the viewpoint that semiconductors or electric materials to be sealed are hardly damaged.

[0149]

The filler may be surface-treated where appropriate. The surface treatment includes alkylation treatment,
10 trimethylsilylation treatment, silicone treatment, treatment with a coupling agent, etc.

[0150]

As examples of the above coupling agent, there may be mentioned a silane coupling agent. The silane coupling agent
15 is not particularly restricted but may be any of those compounds which have, within the molecule, at least one functional group reactive with an organic group and at least one hydrolyzable silyl group. The functional group reactive with an organic group is preferably at least one functional group selected from
20 among epoxy, methacryl, acryl, isocyanate, isocyanurate, vinyl and carbamate groups from the handleability viewpoint. From the viewpoint of curability and adhesive properties, epoxy, methacryl and acryl groups are particularly preferred. The hydrolyzable silyl group is preferably an alkoxysilyl group and,
25 from the reactivity viewpoint, a methoxysilyl and an ethoxysilyl groups are particularly preferred.

[0151]

As preferred examples of the silane coupling agent, there may be mentioned epoxy functional group-containing
30 alkoxysilanes such as 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropyltriethoxysilane, 2-(3,4-epoxycyclohexyl)ethyltrimethoxysilane and 2-(3,4-epoxycyclohexyl)ethyltriethoxysilane; and methacryl or acryl group-containing alkoxysilanes such as
35 3-methacryloxypropyltrimethoxysilane,

3-methacryloxypropyltriethoxysilane,
3-acryloxypropyltrimethoxysilane,
3-acryloxypropyltriethoxysilane,
methacryloxymethyltrimethoxysilane,
5 methacryloxymethyltriethoxysilane,
acryloxymethyltrimethoxysilane and
acryloxymethyltriethoxysilane.

[0152]

In addition, a process for incorporating a filler may be
10 mentioned. There may also be mentioned, for example, a method
of forming a filler in the curable composition which comprises
adding a hydrolyzable silane monomer or oligomer, such as an
alkoxysilane, acyloxysilane or halogenated silane, or an
alkoxide, acyloxide or halide of a metal, such as titanium or
15 aluminum, to the composition of the invention and allowing the
filler formation reaction to proceed in the composition or the
partial reaction product derived from the composition.

[0153]

Among the above fillers, a silica type filler is preferred
20 from the viewpoint that it makes it difficult for the curing
reaction to be inhibited and is highly effective in reducing
the coefficient of linear expansion.

[0154]

The above filler preferably has an average particle
25 diameter of not more than 10 μm , more preferably not more than
5 μm , since good permeability to narrow gaps of a sealing member
can be easily obtained then.

[0155]

From the better permeability to narrow gaps of a sealing
30 member viewpoint, the proportion of particles having a diameter
of 50 μm or larger in the filler is preferably not more than
1% by weight, more preferably not more than 0.1% by weight.

[0156]

The particle diameter distribution of the above filler
35 can be selected in various ways, typically in those ways

employed in and/or proposed for the fillers in the conventional sealing members such as the epoxy type ones. For example, particles larger than 24 μm may amount to not less than 15% by weight while particles not larger than 1 μm may amount to not less than 3% by weight.

[0157]

The average particle diameter of the filler and the proportion of particles not smaller than 50 μm in the filler can be determined using a laser microtrack particle size analyzer.

[0158]

The specific surface area of the above filler can be selected at various levels, typically at the levels employed and/or proposed for the fillers in the conventional sealants, such as the epoxy-based ones. For example, levels of not less than 4 m^2/g , not more than 4 m^2/g , not more than 10 m^2/g and so forth may be optionally employed.

[0159]

The specific surface area can be measured using a BET method-based monosorb specific surface area measuring apparatus.

[0160]

The vitrification percentage of the above filler can be selected at various levels, typically at the levels employed and/or proposed for the fillers in the conventional sealing members, such as the epoxy-based ones. For example, levels of not less than 97%, and the like can be optionally employed.

[0161]

The above filler preferably has a spherical shape from the viewpoint that the sealing member viscosity tends to become low.

[0162]

The filler may comprise one single species or a combination of two or more species.

[0163]

The level of addition of the above filler is not particularly restricted but, from the viewpoint that the linear expansion coefficient reducing effect is high and the composition has good fluidity, the lower limit to the addition level is preferably 30% by weight, more preferably 50% by weight, of the whole composition, and the upper limit to the addition level is preferably 80% by weight, more preferably 70% by weight, of the whole composition.

[0164]

10 Various methods can be employed for filler incorporation. For better storage stability of the intermediate material of the composition, the method comprising mixing the component (B) with a mixture prepared by mixing the component (A) with the component (C) and the filler is preferred. When the component
15 (A) is mixed with a mixture prepared by mixing the component (B) with the component (C) and/or the filler, the composition may undergo denaturation during storage, etc. since the component (B) has reactivity with the moisture in the environment and/or with the filler in the presence and/or
20 absence of the component (C).

[0165]

An antioxidant may be added to the curable composition of the invention. The antioxidant includes those in conventional use, for example citric acid, phosphoric acid,
25 sulfur-containing antioxidants, etc. As the sulfur-containing antioxidants, there may be mentioned mercaptans, mercaptan salts, sulfide carboxylic acid esters, hindered phenol type sulfides and other sulfides, polysulfides, dithiocarboxylic acid salts, thioureas, thiophosphates,
30 sulfonium compounds, thioaldehydes, thioketones, mercaptals, mercaptols, monothio acids, polythio acids, thioamides, sulfoxides, etc.

[0166]

The above antioxidants may be used singly or two or more
35 of them may be used in combination.

[0167]

A radical inhibitor may be added to the composition of the invention. The radical inhibitor includes, for example, phenolic radical inhibitors such as

- 5 2,6-di-tert-butyl-3-methylphenol (BHT),
2,2'-methylenebis(4-methyl-6-tert-butylphenol) and
tetrakis(methylene-3-(3,5-di-tert-butyl-4-hydroxyphenyl)pro
pionate)methane, amine type radical inhibitors such as
phenyl- β -naphthylamine, α -naphthylamine,
10 N,N'-sec-butyl-p-phenylenediamine, phenothiazine and
N,N'-diphenyl-p-phenylenediamine, etc.

[0168]

The above radical inhibitors may be used singly or two or more of them may be used in combination.

15 [0169]

An ultraviolet absorber may be added to the composition of the invention. The ultraviolet absorber includes, for example,

- 2-(2'-hydroxy-3',5'-di-tert-butylphenyl)benzotriazole,
20 bis(2,2,6,6-tetramethyl-4-piperidine) sebacate, and so on.

[0170]

The above ultraviolet absorbers may be used singly or two or more of them may be used in combination.

[0171]

- 25 To the composition of the invention, other than the above components, there may further be added components typically at the levels employed and/or proposed for the fillers in the conventional sealants, such as the epoxy-based ones, and colorants, mold release agents, flame retardants, flame
30 retardant auxiliaries, surfactants, antifoaming agents, emulsifiers, leveling agents, cissing preventing agents, ion trapping agents, thixotropic agents, tackifiers, storage stability improving agents, antiozonants, light stabilizers, thickening agents, plasticizers, reactive diluents,
35 antioxidants, heat stabilizers, electric conductivity

providing agents, antistatic agents, antiradiation agents, nucleating agents, phosphorus-containing peroxide decomposers, lubricants, pigments, metal inactivators, thermal conductivity providing agents, physical property modifiers and so forth, each within the range within which the object and effect of the invention will not be defeated or weakened.

[0172]

The composition of the invention can also be used in the form of a solution in a solvent. The solvent that can be used is not particularly restricted but, for example, there may be specifically mentioned hydrocarbon solvents such as benzene, toluene, hexane and heptane; ether solvents such as tetrahydrofuran, 1,4-dioxane, 1,3-dioxolane and diethyl ether; ketone solvents such as acetone, methyl ethyl ketone and methyl isobutyl ketone; and halogenated solvents such as chloroform, methylene chloride and 1,2-dichloroethane.

[0173]

Among these, preferred are toluene, tetrahydrofuran, 1,3-dioxolane, and chloroform.

[0174]

The amount of the solvent to be used can be selected optionally. However, the lower limit to the amount to be used is preferably 0.1 mL, and the upper limit thereto is preferably 10 mL, per gram of the curable composition. When the solvent amount used is small, the effects of the use of a solvent, for example the viscosity reducing effect and the like effect, tends to become difficult to obtain and, when it is large, the solvent tends to remain in the material, readily causing heat crack and other problems, and tends to become disadvantageous from the cost viewpoint, thus the industrial utility value becomes decrease.

[0175]

The above solvents may be used singly or in the form of a mixed solvent composed of two or more species.

[0176]

The composition of the invention may be incorporated with an additive for improving various properties of the light-emitting diode. As the above additive, for example, there may be mentioned cerium-activated yttrium aluminum garnet type fluorescent substances which absorb the light from the light emitting element and emit a longer wavelength fluorescent light, and the like fluorescent substances; colorants such as a bluing agent which absorb a specific wavelength; inorganic or organic diffusing agents for diffusing the light such as titanium oxide, aluminum oxide, silica, silicon oxide e.g. silica glass, talc, calcium carbonate, melamine resins, CTU guanamine resins and benzoguanamine resins; thermally conductive fillers such as glass, metal oxide e.g. aluminosilicate, etc. and metal nitrides i.e. aluminum nitride, boron nitride, etc.; etc.

[0177]

The additive for improving the light-emitting diode properties may be uniformly contained or may be contained in a gradient manner in the composition. The above filler-containing resin part can be formed as a sealing member of the back side of the light emitting surface by pouring a resin for a sealing member of the front side of the light emitting surface into a mold, and successively pouring a resin containing the above filler. Moreover, the above filler-containing resin part can also be formed by covering a lead terminal by taping both sides after forming a sealing member, and in this condition, immersing the whole lead frame in a tank filled with the above filler-containing resin in such manner that the lower half of the sealing member of the light-emitting diode being immersed, withdrawing thereof, and drying.

[0178]

A light-emitting diode can be produced using the composition of the invention. The light-emitting diode of the invention can be produced by coating the light emitting element with the composition mentioned above.

[0179]

The above light emitting element is not particularly restricted, and may be any of the known light emitting elements which can be used in the light-emitting diodes. For example, there may be mentioned those produced by layering a semiconductor material, by any of various methods, for example the MOCVD, HDVPE, and liquid phase growth methods, on a substrate, if necessary provided with a buffer layer such as a GaN, AlN layer, etc.

As the above substrate, various materials can be used, for example sapphire, spinel, SiC, Si, ZnO, GaN single crystals, etc. Among these, the use of sapphire is preferred since GaN with good crystallinity can be formed with ease and the industrial utility is high.

[0180]

The above semiconductor material to be layered includes, for example, GaAs, GaP, GaAlAs, GaAsP, AlGaInP, GaN, InN, AlN, InGaN, InGaAlN, SiC, etc. Among these, nitride compound semiconductors ($\text{In}_x\text{Ga}_y\text{Al}_z\text{N}$) are preferred from the high brightness viewpoint. The above materials may contain an activator, etc.

[0181]

As the structure of the above light emitting element, there may be mentioned, for example, the homojunction, heterojunction, and double-hetero structure having a MIS junction, pn junction or PIN junction. The single or multiple quantum well structure may also be employed.

[0182]

The light emitting element may have a passivation layer or have no passivation layer.

[0183]

The light emitting element can be provided with electrodes by any of the methods known in the art.

[0184]

The electrodes on the light emitting element can be

electrically connected with lead terminals and so forth by various methods. As the electrically connecting members, preferably ones showing good ohmic and mechanical connectivity with the electrodes of the light emitting element, etc., for example bonding wires made of gold, silver, copper, platinum, aluminum, or alloys thereof, etc. Conductive adhesives or the like composed of a conductive filler, such as silver or carbon, and of a resin filled therewith may also be used. From the good workability viewpoint, aluminum wires or gold wires are preferably used among them.

[0185]

A light emitting element can be thus obtained. In the present invention, any light emitting element showing luminous intensity in the perpendicular direction is not less than 1 cd may be used. When the light emitting element has a luminous intensity in the perpendicular direction of not less than 2 cd, however, the effects of the invention can be produced more significantly and, when the luminous intensity is not less than 3 cd, the effects of the invention are still more significant.

[0186]

The emission output of the above light emitting element is not particularly restricted, and any of those may be used. When the light emitting element shows an output of not less than 1 mW at 20 mA, however, the effects of the invention are significant. When the light emitting element shows an output of not less than 4 mW at 20 mA, the effects of the invention are more significant and, when the light emitting element shows an output of not less than 5 mW at 20 mA, the effects of the invention are still more significant.

[0187]

The above light emitting element that can be used is not particularly restricted, but may be diverse in emission wavelength range, from the ultraviolet to the infrared region. When the element shows a main emission peak wavelength of not longer than 550 nm, however, the effects of the invention are

significant.

[0188]

Only one of the above light emitting element may be used for monochromic light emission, or a plurality of such elements
5 may be used for monochromic or polychromic light emission.

[0189]

The lead terminals to be used in the light-emitting diode of the invention are preferably ones showing good adhesion to electric connection members, such as bonding wires, good
10 electric conductivity, etc. Thus, the lead terminals preferably have an electric resistance of not more than 300 $\mu\Omega\cdot\text{cm}$, more preferably not more than 3 $\mu\Omega\cdot\text{cm}$. As the material of these lead terminals, there may be mentioned, for example, iron, copper, iron-containing copper, tin-containing copper
15 and, further, the products of plating of these with silver, nickel, or the like, etc. The glossiness of these lead terminals may be appropriately adjusted for attaining good spreading of light.

[0190]

20 The light-emitting diode of the invention can be produced by coating the light emitting element with the composition mentioned above. The above-mentioned "coating" includes, within the meaning thereof, not only direct sealing of the light emitting element but also indirect coating. Specifically, the
25 above light emitting element may be directly sealed with the composition of the invention by various methods conventionally in use, or after sealing the light emitting element with known sealing resins such as epoxy resins, silicone resins, acrylic resins, urea resins and imide resins, or glass, and then the
30 above or surround the resultant may be coated with the composition of the invention. Moreover, after sealing the above light emitting element with the composition of the invention, the resultant may be molded with known epoxy resins, silicone resins, acrylic resins, urea resins, imide resins, etc.
35 By these methods, utilizing differences of reflective index and

specific gravity, various effects such as lens effect can be provided.

[0191]

Various methods can be applied as the method of sealing.

5 For example, a liquid composition may be poured into a cup, cavity, package hollow, etc., with the light emitting element disposed on the bottom thereof in advance, using a dispenser or by any other method, followed by curing by heating, etc. It is also possible to allowing a solid composition or highly

10 viscous liquid composition to flow by heating, etc., pouring the same into a package hollow or the like in the same manner and causing curing by heating, etc. The above package can be produced using any of various materials, and there may be mentioned, for example, polycarbonate resins, polyphenylene

15 sulfide resins, epoxy resins, acrylic resins, silicone resins, ABS resins, polybutylene terephthalate resins, polyphthalamide resins, etc. Moreover, a method comprising pouring the composition into a molding form in advance, immersing a lead frame or the like fixed with the light emitting

20 element thereto, and then curing the composition can also be applied. Or the sealing layer comprising the composition may be molded and cured by pouring using a dispenser into a form inserted with the light emitting element, transfer molding, or injection molding, etc. It is also possible to simply cure the

25 composition in a liquid or flowage form by dropwise addition in the shape of the light emitting element or coating. The curable composition can also be molded and cured by mimeographing, screen printing, application via a mask, and the like. Or a method comprising fixing a composition partially

30 cured or cured in a plate-like shape or lens-like shape on the light emitting element can also be applied. Moreover, the composition can be used as a die-bonding agent for fixing the light emitting element to a lead terminal or a package, or as a passivation layer on the light emitting element. Further,

35 it can also be used as a package substrate.

[0192]

The shape of the coated portion is not particularly restricted but may be diverse. For example, there may be mentioned lens-like shapes, plate-like shapes, thin film shapes, those shapes described in Japanese Kokai Publication Hei-06-244458, and the like shapes. These shapes may be formed by molding and curing of a composition or by curing of a composition, followed by after-working.

[0193]

10 The light-emitting diode of the invention can be applied in various types. For example, either types such as lump type, SMD type, and tip type can be applied. As a packaging substrate of SMD and tip types, various ones may be used, and there may be mentioned, for example, epoxy resins, BT resins, ceramics,
15 etc.

[0194]

Further, various known modes can be applied to the light-emitting diode of the invention. For example, there may be mentioned the mode in which a light-reflecting or
20 light-collecting layer is provided on the back of the light emitting element, the mode in which a complementarily colored portion is provided on the bottom to cope with sealant resin yellowing, the mode in which a thin film capable of absorbing shorter wavelength light than the main emission peak is disposed
25 on the light emitting element, the mode in which the light emitting element is sealed with a soft or liquid sealing member and then the circumference is molded with a hard material, the mode in which the light emitting element is sealed with a material containing a fluorescent substance absorbing the light
30 from the light emitting element and emitting a longer wavelength fluorescent light and the circumference is then molded, the mode in which a material containing a fluorescent substance is shaped in advance and then molded together with the light emitting element, the mode in which a sealing member is formed into a
35 special shape to thereby increase the emission efficiency, as

described in Japanese Kokai Publication Hei-06-244458, the mode in which the package is provided with a two-stage hollow to reduce the unevenness of brightness, the mode in which the light-emitting diode is immobilized by insertion into a through
5 hole, the mode in which a thin film capable of absorbing a shorter wavelength light than the main emission wavelength is formed on the light emitting element surface, the mode in which the light emitting element is connected with lead members and the like by flip chip bonding or the like using solder bumps, etc.,
10 for taking out light in the direction of the substrate and the like modes.

[0195]

The light-emitting diode of the invention can be used in various fields of use known in the art. Specifically, such uses
15 include backlights, illumination, light sources for sensors, light sources for gauges in vehicles, signal lamps, display lamps, display devices, light sources for planar light emitters, displays, decorations, various lights, etc.

[0196]

20 The curable composition of the invention is applicable to various optical materials. The optical materials in the invention refer to materials in general used for transmitting light therein such as visible light, infrared rays, ultraviolet rays, X-rays, and laser.

25 [0197]

The optical materials in the invention refer to materials in general used for transmitting light therein such as visible light, infrared rays, ultraviolet rays, X-rays, and laser.

[0198]

30 There may be mentioned, for example, materials used for liquid crystal display devices such as a color filter overcoat, TFT flattening film, and substrate material; and materials used for light-emitting diodes (LED) such as a sealing member and die-bonding agent. In addition, the sealing member in this
35 specification is a concept including a molding agent or a

sealant.

[0199]

There may also be mentioned a substrate material, optical waveguide, prism sheet, deflection plate, wave plate, viewing
5 angle correction film, polarizer protection film, a color filter, etc. used in liquid crystal display fields; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0200]

10 There may also be mentioned a sealing member of LED elements, sealing member of LED, protection film of front face glass, and alternate material for front face glass used in LED display devices; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

15 [0201]

There may also be mentioned an antireflective film, optical correction film, housing material, protection film of front face glass, and alternate material for front face glass used in color PDPs (plasma display); and various coating agents,
20 protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a substrate material, optical waveguide, prism sheet, deflection plate, wave plate, viewing angle correction film, and polarizer protection film in plasma address liquid crystal (PALC) displays; and various coating
25 agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a protection film of front face glass, and alternate material for front face glass in organic electro luminescence displays; and various coating agents, protection films, sealing members, adhesives, etc. used
30 for them. There may also be mentioned various film substrates, a protection film of front face glass, and alternate material for front face glass in field emission displays (FED); and various coating agents, protection films, sealing members, adhesives, etc. used for them.

35 [0202]

In the optical record fields, there may be mentioned VD (video disk), CD/CD-ROM, CD-R/RW, DVD-R/DVD-RAM, MO/MD, PD (phase change disk), disk substrate material for optical cards, pickup lens, and protection film; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0203]

In the optical apparatus fields, there may be mentioned a lens material for steel cameras, finder prism, target prism, finder cover, and light-sensitive sensor part; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a photographing lens for video cameras, and finder; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a projection lens and protection film for projection TVs; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a lens material and various films for optical sensing apparatus; and various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0204]

In the optical part fields, there may be mentioned a fiber material for the peripheral of optical switch, lens, waveguide, element in optical communication systems; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned an optical fiber material and ferrule for the peripheral of optical connectors; and various coating agents, protection films, sealing members, adhesives, etc. used for them. As optical passive parts and optical circuit parts, there may be mentioned a lens and waveguide; and various coating agents, protection films, sealing members, adhesives, etc. used for them. There may also be mentioned a substrate material and fiber material for the periphery of opto-electronic integrated circuit (OEIC); and

various coating agents, protection films, sealing members, adhesives, etc. used for them.

[0205]

5 In the optical fiber fields, there may be mentioned
sensors for industrial application such as lightning for
ornament displays and light guide, optical fibers for
connecting communication infrastructure and domestic digital
equipment such as a display and indicator; and various coating
agents, protection films, sealing members, adhesives, etc. used
10 for them.

[0206]

In the semiconductor integrated circuit peripheral
materials, there may be mentioned resist a material of micro
lithography for LSI and super LSI materials.

15 [0207]

In the car and transport plane fields, there may be
mentioned lump materials such as for headlight, taillight and
indoor lamp for cars, a lamp reflector, a lamp lens, various
interior and exterior products such as an exterior and interior
20 panel, and glass substitute; and various coating agents,
protection films, sealing members, adhesives, etc. used for
them. There may also be mentioned an exterior part and glass
substitute for railway vehicles; and various coating agents,
protection films, sealing members, adhesives, etc. used for
25 them. There may also be mentioned an exterior part and glass
substitute for airplanes; and various coating agents,
protection films, sealing members, adhesives, etc. used for
them.

[0208]

30 In the architectural fields, there may be mentioned a
glass intermediate film, glass substitute, and solar cell
peripheral material; and various coating agents, protection
films, sealing members, adhesives, etc. used for them.

[0209]

35 For agriculture, there may be mentioned a coating film

for green houses.

[0210]

As the optical and electric functional organic materials for the next generation, there may be mentioned an organic EL
5 element peripheral material, organic photorefractive element, optical amplification element which is a light-light conversion device, optical calculation element, substrate material for the peripheral of organic solar cells, fiber material, sealing member of element; and various coating agents, protection films,
10 sealing members, adhesives, etc. used for them.

[0211]

As a process for curing the curable composition of the invention, the reaction may be allowed to proceed by mere mixing or by heating. From the viewpoint that the reaction rate is
15 rapid and materials generally high in heat resistance are readily obtainable, the method comprising heating to allow the reaction to proceed is preferred.

[0212]

The reaction temperature may be selected at various
20 levels. However, the usable temperature is 30 to 300°C, more preferably 100 to 250°C, still more preferably 150 to 200°C. At lower reaction temperatures, the reaction time for the reaction to proceed to a satisfactory extent becomes long, whereas at higher temperatures, molding process tends to be
25 difficult.

[0213]

The reaction may be carried out at a constant temperature, or the temperature may be varied stepwise or continuously according to need. The reaction is preferably carried out with
30 the temperature varied stepwise or continuously, not at a constant temperature, because the cured product to be obtained tends to be strain-free and uniform.

[0214]

The reaction time can also be selected at various levels.
35 The reaction is preferably carried out at lower temperature for

comparatively long time, not at elevated temperature for comparatively short time, because the cured product to be obtained tends to be strain-free and uniform.

[0215]

5 The pressure during reaction also can be selected at various levels according to need. Thus, the reaction can be carried out at ordinary pressure, at an elevated pressure or under reduced pressure. The reaction is preferably carried out under reduced pressure because vaporous components to be
10 produced are easily eliminated by hydrolysis condensation.

[0216]

 The shapes of the optical material obtained by curing is not particularly restricted since various shapes are applicable according to its application, and include, for example, film,
15 sheet, tube, rod, coat film, and bulk shapes.

[0217]

 Molding methods also include various methods including conventional methods for molding thermoplastic resins. For example, molding methods such as casting method, press method,
20 cast molding method, transfer molding method, coating method, and RIM method can be applied. As molding tools, polished glass, hard stainless polished board, polycarbonate board, polyethylene terephthalate board, polymethyl methacrylate board, etc. can be applied. Moreover, for improving
25 mold-releasing ability with the molding tool, it is possible to use a polyethylene terephthalate film, polycarbonate film, polyvinyl chloride film, polyethylene film, polytetrafluoroethylene film, polypropylene film, polyimide film, etc.

30 [0218]

 In the molding, various treatments may be applied according to need. For example, such treatments can also be applied as one comprising defoaming the composition or partly-reacted composition by centrifugation, reduced
35 pressure, etc. for inhibiting generation of a void in the step

of the molding, or one comprising releasing pressure once in the step of pressing.

[0219]

[Examples]

5 The following examples and comparative examples illustrate the present invention. They are, however, by no means limitative of the scope of the present invention.

(Synthesis Example 1)

10 A 5-L separable flask was charged with 1.8 kg of toluene and 1.44 kg of 1,3,5,7-tetramethylcyclotetrasiloxane, and heated until the inside temperature arrived at 104°C. Thereto was added dropwise a mixture of 200 g of triallyl isocyanurate, 1.44 mL of platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) and 200 g of toluene. The
15 resulting mixture was heated under reflux in an oil bath at 120°C for 7 hours. Then, 1.7 g of 1-ethynyl-1-cyclohexanol was added. The unreacted portion of 1,3,5,7-tetramethylcyclotetrasiloxane and the toluene were distilled off under reduced pressure. ¹H-NMR revealed that the
20 product was the reaction product resulting from part of the SiH groups of 1,3,5,7-tetramethylcyclotetrasiloxane with triallyl isocyanurate (hereinafter referred to as partial reaction product A, SiH value: 8.2 mmol/g, allyl value: 0.12 mmol/g).

25 (Example 1)

 A mixture (mixture A) was prepared by mixing up triallyl isocyanurate (12.04 g), 90 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum), and 150 mg of trimethyl borate. B mixture (mixture B) was
30 prepared by mixing up 17.96 g of the partial reaction product (A) prepared in Synthesis Example 1, 90 mg of 1-ethynyl-1-cyclohexanol, 750 mg of γ-glycidoxypropyltrimethoxysilane. After mixing up the above mixtures A and B with stirring and degassing, the mixture was
35 poured into a cell produced by inserting a 3-mm-thick silicone

rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour and 150°C for 1 hour, in that order, to obtain a transparent and hard molding.

5

(Example 2)

A mixture (mixture A) was prepared by mixing up triallyl isocyanurate (12.04 g), 90 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum),
10 and 150 mg of tributyl borate. B mixture (mixture B) was prepared by mixing up 17.96 g of the partial reaction product (A) prepared in Synthesis Example 1, 90 mg of 1-ethynyl-1-cyclohexanol, 750 mg of γ -glycidoxypropyltrimethoxysilane. After mixing up the above
15 mixtures A and B with stirring and degassing, the mixture was poured into a cell produced by inserting a 3-mm-thick silicone rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour and 150°C for 1 hour, in that order,
20 to obtain a transparent and hard molding.

(Example 3)

A mixture (mixture A) was prepared by mixing up triallyl isocyanurate (12.04 g), 90 mg of a platinum-vinylsiloxane
25 complex solution in xylene (containing 3% by weight of platinum), and 150 mg of triisopropyl borate. B mixture (mixture B) was prepared by mixing up 17.96 g of the partial reaction product (A) prepared in Synthesis Example 1, 90 mg of 1-ethynyl-1-cyclohexanol, 750 mg of
30 γ -glycidoxypropyltrimethoxysilane. After mixing up the above mixtures A and B with stirring and degassing, the mixture was poured into a cell produced by inserting a 3-mm-thick silicone rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for
35 1 hour, 120°C for 1 hour and 150°C for 1 hour, in that order,

to obtain a transparent and hard molding.

(Example 4)

5 A mixture (mixture A) was prepared by mixing up triallyl
isocyanurate (12.04 g), 90 mg of a platinum-vinylsiloxane
complex solution in xylene (containing 3% by weight of platinum),
and 150 mg of trinormalpropyl borate. B mixture (mixture B)
was prepared by mixing up 17.96 g of the partial reaction product
(A) prepared in Synthesis Example 1, 90 mg of
10 1-ethynyl-1-cyclohexanol, 750 mg of
 γ -glycidoxypropyltrimethoxysilane. After mixing up the above
mixtures A and B with stirring and degassing, the mixture was
poured into a cell produced by inserting a 3-mm-thick silicone
rubber sheet as a spacer between two glass plates, heated in
15 a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for
1 hour, 120°C for 1 hour and 150°C for 1 hour, in that order,
to obtain a transparent and hard molding.

(Comparative Example 1)

20 A mixture (mixture A) was prepared by mixing up triallyl
isocyanurate (12.04 g), 90 mg of a platinum-vinylsiloxane
complex solution in xylene (containing 3% by weight of platinum),
and 150 mg of ALCH-TR (Kawaken Fine Chemicals Co., Ltd.). B
mixture (mixture B) was prepared by mixing up 17.96 g of the
25 partial reaction product (A) prepared in Synthesis Example 1,
90 mg of 1-ethynyl-1-cyclohexanol, 750 mg of
 γ -glycidoxypropyltrimethoxysilane. After mixing up the above
mixtures A and B with stirring and degassing, the mixture was
poured into a cell produced by inserting a 3-mm-thick silicone
30 rubber sheet as a spacer between two glass plates, heated in
a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for
1 hour, 120°C for 1 hour and 150°C for 1 hour, in that order,
to obtain a transparent and hard molding.

35 (Measurement Example 1)

For the samples obtained in Example 1 and Comparative Example 1, tensile elasticity and tensile elongation were evaluated. The test samples were prepared as follows. The plate-shaped curing products of 3 mm in thickness were cut into rectangles of 6 x 55 x 3 mm, and near the center of longitudinal sides thereof were notched from both sides in U shape of $r=1.0$ mm and depth of 1 mm. The measurement was carried out using an autograph placed under the condition of 50% RH at 23°C, and the distance between the chucks was set at 15 mm. The U-notch parts of the test samples were placed near the center of the chucks, both ends thereof were pinched with the chucks, and tensile test was carried out at a tensile speed of 1 mm/min. The obtained results are shown in the following Table.

[0220]

15 [Table 1]

	Tensile characteristics	
	strength, MPa	elongation, %
Example 1	30	6
Comper.Ex.1	21	3

20

From Table 1, it is found that the curing product of the invention has high tensile strength and elongation.

25 (Measurement Example 2)

For the samples obtained in Examples 1 to 4 and Comparative Example 1, the light transmittance at a wavelength of 470 nm was evaluated at the initial stage, after heating at 180°C for 24 hours, 190°C for 24 hours, and 200°C for 24 hours. A spectrophotometer U-3300 manufactured by Hitachi, Ltd. was used as measurement equipment. The light transmittance of the curing product obtained in the Examples and Comparative Examples are shown below.

[0221]

35 [Table 2]

	Light transmittance at 470 nm, %			
	Initial stage	180°C/24h	190°C/24h	200°C/24h
Example 1	90	82	79	72
Example 2	89	82	65	52
Example 3	90	82	66	52
Example 4	89	81	61	51
Comper.Ex.1	88	34	18	11

From Table 2, it is found that the curing products of the invention have small changes in the light transmittance at high temperatures.

(Measurement Example 3)

To resin-made packages of 2.5 mm in width, 3 mm in length, and 2 mm in depth and having lead electrodes, the curable compositions of Examples 1 to 4 and Comparative Example 1 were filled. The packages were heated at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for 1 hour and 150°C for 1 hour, in that order, and subjected to a thermal shock test. The thermal shock test was carried out by repeating the following cycle for 100 times: exposing the packages to -40°C for 5 minutes, heating to 100°C, exposing to 100°C for 5 minutes, cooling to -40°C, and exposing to -40°C for 5 minutes. After completion of 100 cycles, the packages sealed with the curing products of Examples showed preferable adhesive properties with the package bottom and side surfaces, the result was equivalent to that of the curing product of Comparative Example 1.

(Example 5)

The sheet-shaped curing product produced in Example 1 is cut into an appropriate form, and fixed on a light-transmitting window disposed on a metal cap for can type. Meanwhile, a light emitting element having a double-hetero structure in which an Si- and Zn-doped InGaN active layer formed on a sapphire substrate by MOCVD (Metal Organic Chemical Vapor Deposition) method being sandwiched with n- and p-type AlGaIn clad layers

is prepared. Successively, this light emitting element is mounted on a metal stem for can type, and p- and n-electrodes were wire-bonded with an Au line to the respective leads. This is air-sealed with the above metal cap for can type. In this manner, a can type light-emitting diode can be produced.

(Example 6)

On a washed sapphire substrate, by MOCVD (Metal Organic Chemical Vapor Deposition) method, the below-mentioned layers are laminated in the following order: n-type GaN layer which is an undoped nitride semiconductor; GaN layer which is to be an n-type contact layer formed with an Si-doped n-type electrode; n-type GaN layer which is an undoped nitride semiconductor; GaN layer which is to be a barrier layer constituting a light emitting layer; InGaN layer constituting a well layer; GaN layer which is to be a barrier layer (quantum well structure); AlGaN layer as a p-type clad layer doped with Mg on a light emitting layer; and GaN layer which is an Mg-doped p-type contact layer. By etching, the surfaces of p- and n-contact layers are exposed on the same side of the nitride semiconductor on the sapphire substrate. On the respective contact layers, Al is deposited by a sputtering method to form positive and negative electrodes. A scribing line was drawn on the prepared semiconductor wafer, and then the wafer is divided by external pressure to form a light emitting element.

[0222]

On the bottom surface of a silver-plated mount lead cup constituted of iron-containing copper, the above light emitting element is die-bonded using an epoxy resin composition as a die bond resin. The resultant is heated at 170°C for 75 minutes to cure the epoxy resin composition to fix the light emitting element. Next, the positive and negative electrodes of the light emitting element, the mount lead and inner lead are wire-bonded with an Au line to gain electric access.

[0223]

A curable composition prepared in the same manner as Example 1 is poured into a casting case, which is a bombshell-shaped molding form. Parts of the mount lead and inner lead, the cup of which is disposed with the above light emitting element, are inserted into the casting case to carry out an initial curing at 100°C for 1 hour. The light-emitting diode is withdrawn from the casting case and cured at 120°C for 1 hour under nitrogen atmosphere. Thereby, a lump type light-emitting diode such as a bombshell-shaped one can be produced.

(Example 7)

A curable composition and a light emitting element is produced by the method described in Example 1.

[0224]

A pair of copper foil patterns is formed on a glass epoxy resin by etching to produce a substrate having lead electrodes. A light emitting element is die-bonded on the glass epoxy resin using an epoxy resin. The respective electrodes of the light emitting element and respective lead electrodes are wire-bonded with an Au line to gain electric access. On the substrate, a glass epoxy resin having a through hole is fixed and disposed using an epoxy resin to function as a mask and sidewall. In this condition, the resultant is disposed in vacuum equipment, the curable composition is dispensed on the glass epoxy resin substrate disposed with the light emitting element, and the curable composition is filled into a cavity utilizing the through hole. In this condition, the composition is cured at 100°C for 1 hour, and further at 150°C for 1 hour. By dividing with every light-emitting diode tip, a tip type light-emitting diode can be produced.

(Example 8)

A curable composition and a light emitting element is produced by the method described in Example 1.

[0225]

By insert molding, a package of a tip type light-emitting diode is formed using a PPS resin. Inside of the package, an aperture to be disposed with a light emitting element is
5 equipped and a silver-plated copper plate is placed as an external electrode. A light emitting element is fixed using an epoxy resin by die bonding inside the package. An Au line, which is a conductive wire, is electrically connected to the respective electrodes of the light emitting element and the
10 respective external electrodes disposed in the package by wire bonding. Inside of the package aperture, a curable composition is filled as a sealing member. In this condition, the composition is cured at 100°C for 1 hour, and further at 150°C for 1 hour. In this manner, a tip type light-emitting diode
15 can be produced.

(Example 15)

Triallyl isocyanurate (12.04 g) was used as the component (A), 17.96 g of the reaction product (B1) prepared in Synthesis
20 Example 1 as the component (B), 90 mg of a platinum-vinylsiloxane complex solution in xylene (containing 3% by weight of platinum) as the component (C), 750 mg of γ -glycidoxypropyltrimethoxysilane as the component (D), and 150 mg of trinormalbutyl borate as the component (E). A mixture
25 (mixture A) was prepared in advance by mixing up the above components (A), (C) and (E) with stirring. Separately, a mixture (mixture B) was prepared in advance by mixing up the above components (B) and (D), together with 90 mg of 1-ethynyl-1-cyclohexanol with stirring. A one-pot mixture was
30 prepared by mixing up the above mixtures A and B with stirring and degassing. The above one-pot mixture was poured into a cell produced by inserting a 3-mm-thick silicone rubber sheet as a spacer between two glass plates, heated in a hot air drier at 60°C for 6 hours, 70°C for 1 hour, 80°C for 1 hour, 120°C for
35 1 hour, 150°C for 1 hour and 170°C for 30 minutes, in that order,

to obtain a transparent and hard molding.

[0226]

[Effect of the Invention]

5 The curable composition of the invention used for a
light-emitting diode is high in toughness and low in coloration
at high temperatures, it can be used as a sealing member for
light-emitting diodes.

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[Document Name] Abstract

[Abstract]

[Subject] The present invention provides a curable composition providing a curing product having high toughness and low coloration at high temperatures.

[Means for Solving]

A curable composition which contains, as essential components,

(A) an organic backbone-based compound containing at least two carbon-carbon double bonds reactive with a SiH group in each molecule,

(B) a silicon compound having at least two SiH groups in each molecule,

(C) a hydrosilylation catalyst,

(D) a silane coupling agent and/or an epoxy group-containing compound, and

(E) a borate ester.

[Selective Figure] none